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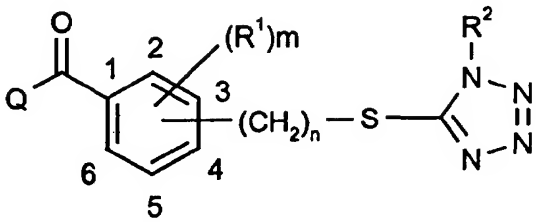
(54) Title: NOVEL TETRAZOLE DERIVATIVES



(I)

(57) Abstract: The instant invention relates to novel tetrazole derivatives of the formula (I) wherein R^1 represents halogen, methyl, ethyl, halomethyl, methoxy, ethoxy, C_{1-2} haloalkoxy, methylthio, ethylthio, C_{1-3} alkylsulfonyl, methylsulfonyloxy, ethylsulfonyloxy, nitro or cyano, R^2 represents C_{1-6} alkyl, C_{3-6} cycloalkyl which may be optionally substituted with halogen or C_{1-3} alkyl, C_{1-4} haloalkyl, C_{2-6} alkenyl, or phenyl which may be optionally substituted with halogen, C_{1-3} alkyl, C1-4 haloalkyl, C_{2-6} alkenyl, or phenyl which may be optionally substituted with halogen, C_{1-3} alkyl, C_{1-2} haloalkyl or nitro, m represents 0, 1 or 2,two R^1 s may be identical or different, in case m represents 2, n represents 1 or 2, Q represents one the cyclic groups which are mentioned in the specification, to intermediates and several processes for their preparation, to their use as herbicides and to novel compositions containing them.

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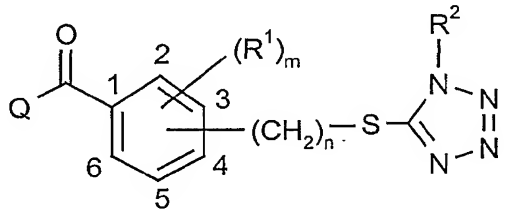
NOVEL TETRAZOLE DERIVATIVES

The present invention relates to novel tetrazole derivatives, to processes for their preparation, to their intermediates, to their use as herbicides and to novel herbicidal compositions for use in paddy fields.

It has been already known that certain kinds of tetrazole derivatives show a herbicidal activity (cf. Japanese Laid-open Patent Application No. 12275/1999, No. 21280/1999 etc.). Furthermore, it has been known that certain kinds of heterocyclic derivatives show a herbicidal activity (cf. U.S. Patent Specifications No. 5834402, No. 5846906, DE-A-19846792, WO 99/10327 etc.).

There have now been found novel tetrazole derivatives of the formula (I)



(I)

wherein

R^1 represents halogen, methyl, ethyl, halomethyl, methoxy, ethoxy, C_{1-2} haloalkoxy, methylthio, ethylthio, C_{1-3} alkylsulfonyl, methylsulfonyloxy, ethylsulfonyloxy, nitro or cyano,

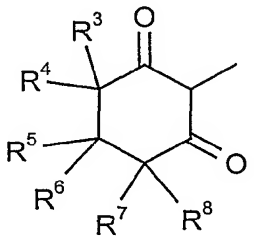
R^2 represents C_{1-6} alkyl or C_{3-6} cycloalkyl which may be optionally substituted with halogen or C_{1-3} alkyl, or represents C_{1-4} haloalkyl, C_{2-6} alkenyl, or phenyl which may be optionally substituted with halogen, C_{1-3} alkyl, C_{1-2} haloalkyl or nitro,

m represents 0, 1 or 2,

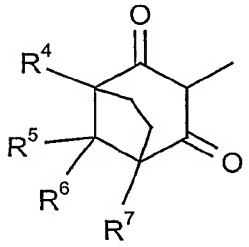
and the two R^1 substituents may be identical or different, in case m represents 2,

n represents 1 or 2,

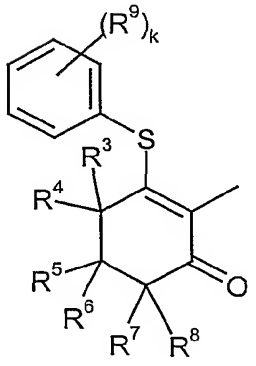
Q represents one of the following groups



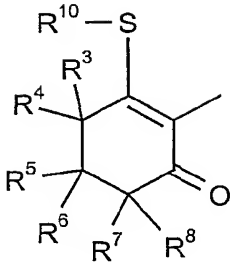
(Q-1),



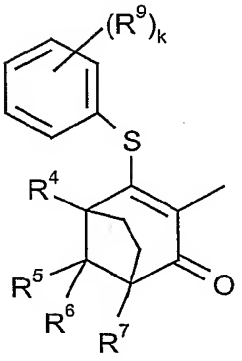
(Q-2),



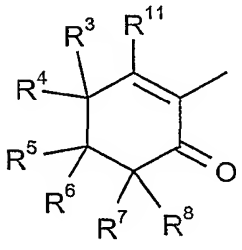
(Q-3),



(Q-4),

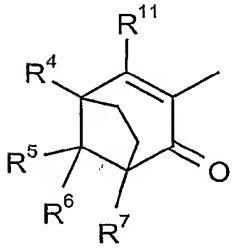


(Q-5),



(Q-6),

or



(Q-7),

wherein

R^3, R^4, R^5, R^6, R^7 and R^8 are identical or different and each represents a hydrogen atom or methyl,

R^9 represents a hydrogen atom, halogen, C_{1-3} alkyl, halomethyl, methoxy or nitro,

R^{10} represents C_{1-6} alkyl,

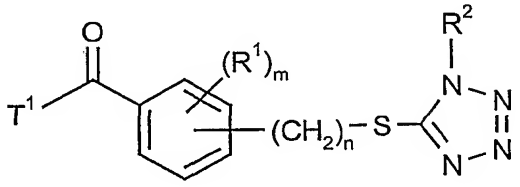
R^{11} represents halogen, and

k represents 1 or 2.

The compounds of the formula (I), according to the invention, can be obtained by a process wherein

a) in case of preparing a compound of the formula (I) wherein Q represents groups (Q-1) or (Q-2):

compounds of the formula (II)

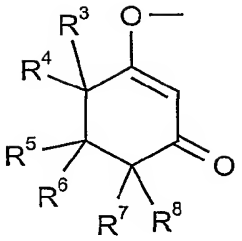


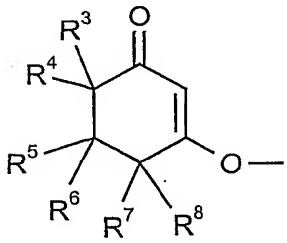
(II)

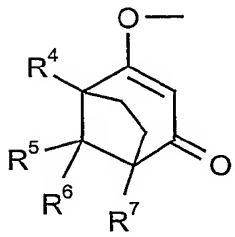
wherein

R^1, R^2, m and n have the same definition as aforementioned, and

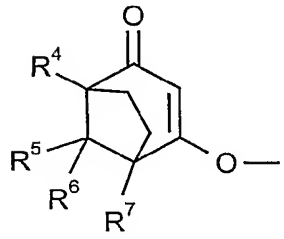
T^1 represents one of the following groups


,


,


,

or


,

wherein

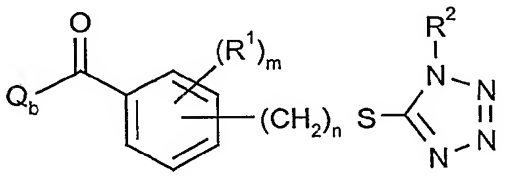
R^3, R^4, R^5, R^6, R^7 and R^8 have the same definition as aforementioned,

are reacted to a rearrangement in the presence of inert solvents, and if appropriate, in the presence of a base and a cyanide, and if appropriate, in the presence of a phase-transfer catalyst,

or

b) in case of preparing a compound of the formula (I) wherein Q represents groups (Q-6) or (Q-7) and R^{11} in said groups represents chloro or bromo:

compounds of the formula (Ib)

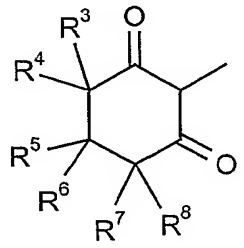


(Ib)

wherein

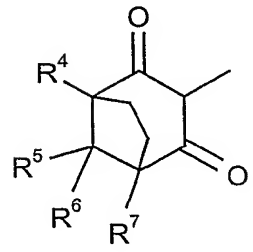
R^1, R^2, m and n have the same definition as aforementioned, and

Q_b represents one of the following groups



(Q-1),

or



(Q-2),

wherein

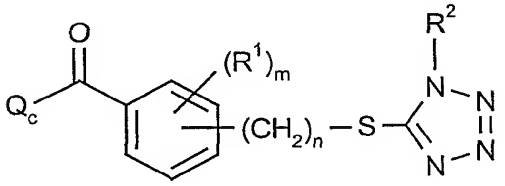
R^3, R^4, R^5, R^6, R^7 and R^8 have the same definition as aforementioned,

are reacted with a halogenating agent in the presence of inert solvents,

or

c) in case of preparing a compound of the formula (I) wherein Q represents groups (Q-3), (Q-4) or (Q-5):

compounds of the formula (Ic)

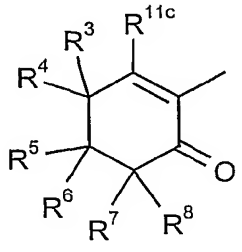


(Ic)

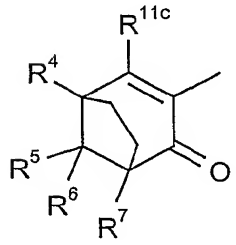
wherein

R^1, R^2, m and n have the same definition as aforementioned, and

Q_c represents one of the following groups



or



wherein

R^3, R^4, R^5, R^6, R^7 and R^8 have the same definition as aforementioned,

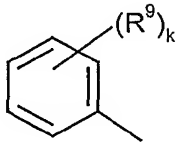
R^{11c} represents chloro or bromo,

are reacted with compounds of the formula (III)

$$R^{12}\text{-SH} \qquad \text{(III)}$$

wherein

R^{12} represents the following group



or

R^{10}

wherein

R^9, R^{10} and k have the same definition as aforementioned,

in the presence of inert solvents, and if appropriate, in the presence of an acid binding agent.

The tetrazole derivatives of the formula (I) provided by the present invention show stronger herbicidal activity than with the compounds described in the aforementioned prior art references.

In the formulae:

"Halogen" represents fluoro, chloro, bromo or iodo, and preferably represents fluoro, chloro or bromo.

"Alkyl" can be straight chain or branched chain and there can be specifically mentioned, for example, methyl, ethyl, n- or iso-propyl, n-, iso-, sec- or tert-butyl, n-, iso-, neo-, or tert-pentyl and n- or iso-hexyl.

"Cycloalkyl" includes cyclopropyl, cyclobutyl, cyclopentyl and cyclohexyl. These cycloalkyls may be optionally substituted with halogen (for example, fluoro, chloro, bromo etc.), C_{1-3} alkyl (for example, methyl, ethyl, n- or iso-propyl etc.) and in case that a plurality of substituents exist, they may be identical or different. As specific examples of such substituted cycloalkyls there can be mentioned 1-methylcyclopropyl, 1-ethylcyclopropyl, 1-n-propylcyclopropyl, 1-methyl-2-fluorocyclopropyl, 2-methylcyclopropyl, 2-fluorocyclopropyl, 1-methyl-2,2-difluorocyclopropyl, 1-methyl-2,2-dichlorocyclopropyl, 2,2difluorocyclopropyl, 2-methylcyclopentyl, 1-methylcyclohexyl, 2-methylcyclohexyl, 3-methylcyclohexyl, 4-methylcyclohexyl, 2,3-dimethylcyclohexyl, 2,6-dimethylcyclohexyl and 2,5-dimethylcyclohexyl.

As "alkenyl" there can be mentioned, for example, vinyl, allyl, 1-methylallyl, 1,1-dimetylallyl and 2-butenyl.

"Haloalkyl" represents straight chain or branched chain alkyl, of which at least one hydrogen is substituted with halogen, and there can be mentioned, for example, C_{1-4} alkyl substituted with 1-6 fluoro and/or chloro, specifically difluoromethyl, trifluoromethyl, 2,2,2-trifluoroethyl, dichloromethyl, 2-chloro-1,1,2-trifluoroethyl, 3-fluoropropyl, 3-chloropropyl, 2,2,3,3,3-pentafluoropropyl and 1,2,2,3,3,3-hexafluoropropyl.

The Haloalkyl part in "haloalkoxy" can have the same definition as the afore-

mentioned "haloalkyl" and as "haloalkoxy" there can be specifically mentioned, for example, difluoromethoxy, trifluoromethoxy, 2-fluoroethoxy, 2-chloroethoxy, 2-bromoethoxy, 2,2,2- trifluoroethoxy and 3-chloropropoxy.

"Alkylsulfonyl" represents an alkyl-SO_2- group, wherein the alkyl part has the above-mentioned meaning, and includes specifically methylsulfonyl, ethylsulfonyl, n- or iso-propylsulfonyl.

As preferred definitions in the formula (I) there can be mentioned:

R^1 preferably represents fluoro, chloro, bromo, methyl, ethyl, trifluoromethyl, methoxy, ethoxy, C_{1-2} haloalkoxy, methylthio, ethylthio, methylsulfonyl, ethylsulfonyl, methylsulfonyloxy, ethylsulfonyloxy, nitro or cyano.

R^2 preferably represents C_{1-3} alkyl, cyclopropyl which may be optionally substituted with fluoro, chloro, methyl or ethyl, C_{1-3} haloalkyl, C_{2-4} alkenyl, or phenyl which may be optionally substituted with fluoro, chloro, methyl, ethyl, trifluoromethyl or nitro.

m preferably represents 1 or 2.

n preferably represents 1 or 2.

R^9 preferably represents a hydrogen atom, fluoro, chloro, methyl, ethyl or trifluoromethyl.

R^{10} preferably represents methyl or ethyl.

R^{11} preferably represents chloro or bromo.

k preferably represents 1.

As more preferred definitions in the formula (I) there can be mentioned:

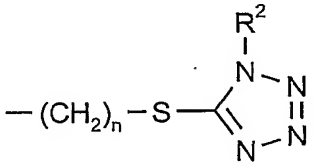
R^1 more preferably represents chloro, bromo, methyl or methylsulfonyl,

R^2 more preferably represents methyl, ethyl, n-propyl, isopropyl or cyclopropyl,

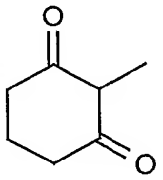
m more preferably represents 2, and in this case the two R^1 substituents are bond respectively to the 2-position and 4-position of a benzene ring and the two R^1 substituents may be identical or different.

n more preferably represents represents 1.

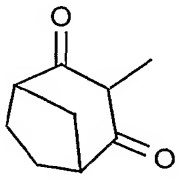
In a most preferred group of the inventive compounds the group



bonds to the 3-position (acccording to formula (I)) of the benzene ring.
In another most preferred group Q represents one of the following groups



or

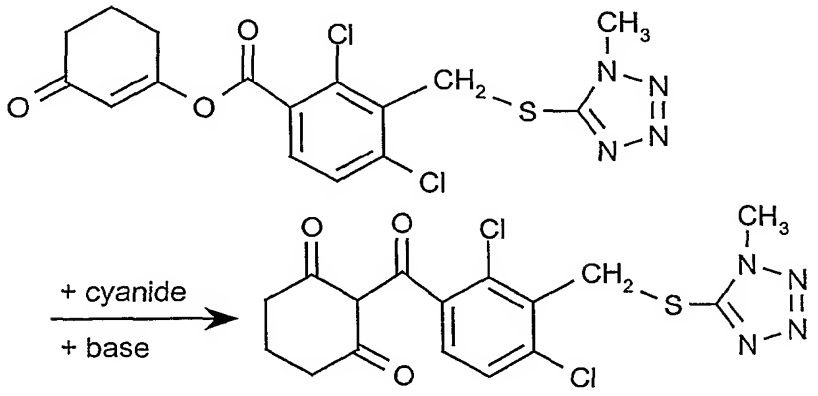


The substituents among the different ranges of preference can be combined without limitation among each other.

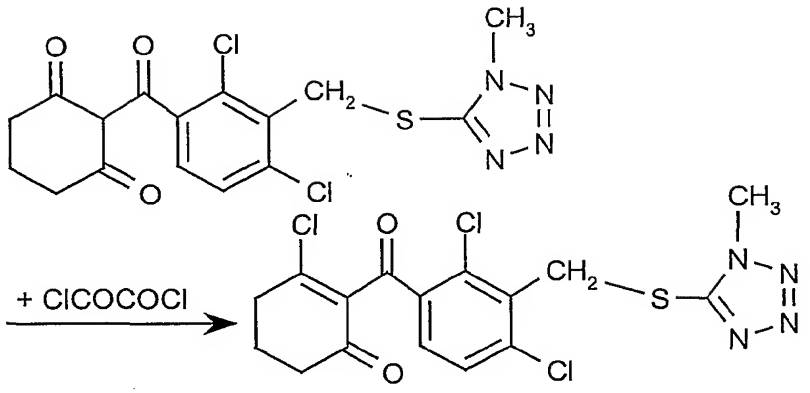
limitation among each other.

However, as a preferred group of compounds there may be explicitly mentioned the compounds of the formula (I) wherein the substituents have the preferred meaning as described above, and as a more preferred group of compounds there may be explicitly mentioned the compounds of the formula (I) wherein the substituents have the more preferred meaning as described above.

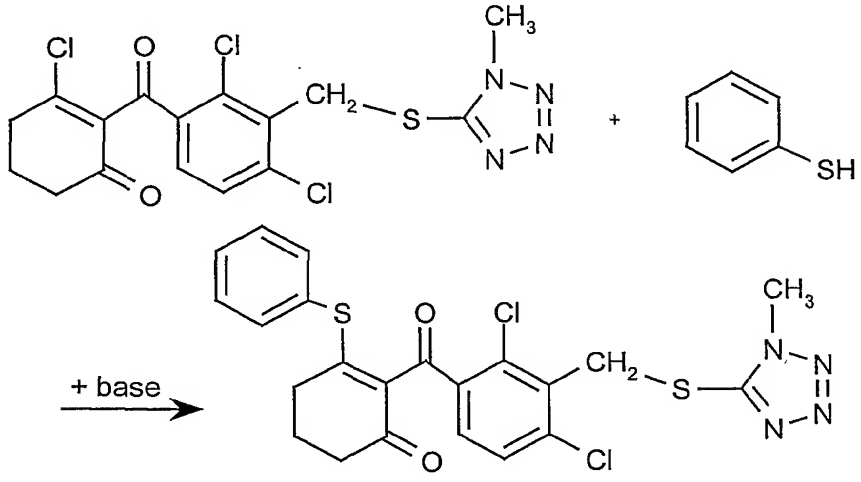
The aforementioned preparation process (a) can be illustrated by the following reaction formula, in case of using, for example, 3-oxo-1-cyclohexenyl 2,4-dichloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoate as the starting material.



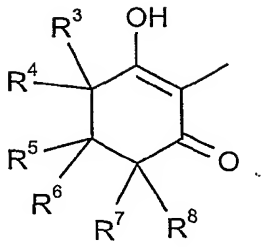
The aforementioned preparation process (b) can be illustrated by the following reaction formula, in case of using, for example, 2-{2,4-dichloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoyl}cyclohexane-1,3-dione as the starting material, and, for example, oxalyl dichloride as chlorinating agent.



The aforementioned preparation process (c) can be illustrated by the following reaction formula, in case of using, for example, 3-chloro-2-{2,4-dichloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoyl}-2-cyclohexen-1-one and thiophenol as the starting materials.

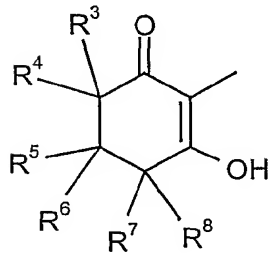


It is further mentioned that the group (Q-1) defined for Q in the above-mentioned formula (I) can also exist in the following two tautomeric forms



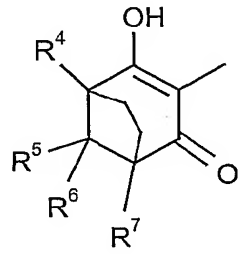
(Q-1a)

or



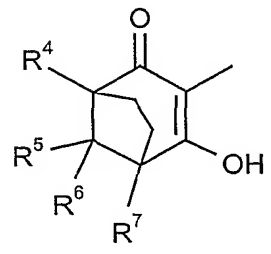
(Q-1b).

It is also mentioned that the group (Q-2) defined for Q in the above-mentioned formula (I) can also exist in the following two tautomeric forms



(Q-2a)

or

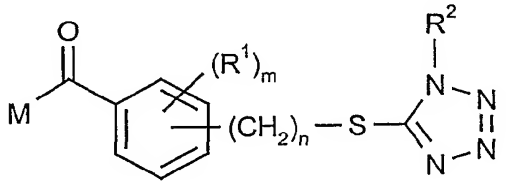


(Q-2b).

Thus, the compounds of the formula (I) of the present invention include the compounds of the formula (I) wherein Q represents the above-mentioned tautomeric groups (Q-1a), (Q-1b), (Q-2a) or (Q-2b) as group Q-1 or Q-2 respectively. In the

present specification, however, it should be understood that these tautomeric groups are represented, unless specified, by the illustration of group (Q-1) or group (Q-2).

The compounds of the formula (II), the starting materials in the above-mentioned preparation process (a), are novel compounds which were not described in the literature up to the present and can be prepared according to the process described in various publications (e.g., Japanese Laid-open Patent Publications No. 222/1990, No. 173/1990, No. 6425/1990 etc.) by reacting compounds of the formula (IV)



(IV)

wherein

R^1, R^2, m and n have the same definition as aforementioned, and

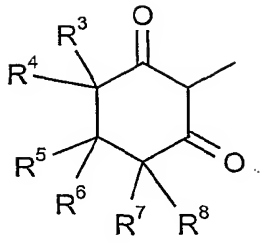
M represents halogen,

with compounds of the formula (V)

Q_a-H (V)

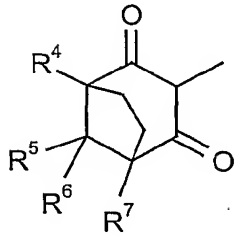
wherein

Q_a represents one of the following groups



(Q-1)

or



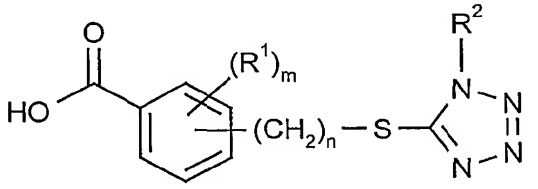
(Q-2)

wherein

R^3, R^4, R^5, R^6, R^7 and R^8 have the same definition as aforementioned,

in an appropriate diluent, for example, dichloromethane, in the presence of an appropriate condensing agent, for example, triethylamine.

The compounds of the formula (IV) used in the above-mentioned reaction are also novel compounds which were not described in the literature up to the present and can be prepared, for example, by reacting compounds of the formula (VI)



(VI)

wherein

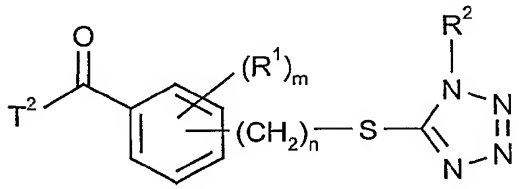
R^1, R^2, m and n have the same definition as aforementioned,

with a halogenating agent, for example, phosphorus oxychloride, phosphorus oxy-

bromide, phosphorus trichloride, phosphorus tribromide, phosgene, oxalyl dichloride, thionyl chloride, thionyl bromide.

The compounds of the formula (V) used as the starting materials in the preparation of the compounds of the above-mentioned formula (II) are per se known and commercially available or can be easily prepared according to the processes described in various publications (e.g., Japanese Laid-open Patent Publications No. 6425/1990, No. 265415/1998, No. 265441/1998).

The compounds of the formula (VI) used for the preparation of the compounds of the above-mentioned formula (IV) are also novel compounds which were not described in the literature up to the present and can be easily prepared, for example, by hydrolyzing compounds of the formula (VII)



(VII)

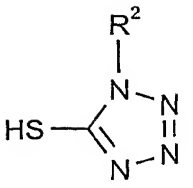
wherein

R^1, R^2, m and n have the same definition as aforementioned, and

T^2 represents C_{1-4} alkoxy, preferably methoxy or ethoxy,

in an appropriate diluent, for example, aqueous dioxane, in the presence of an appropriate base, for example, sodium hydroxide.

The compounds of the above-mentioned formula (VII) are also novel compounds and can be easily obtained, for example, by reacting compounds of the formula (VIII)

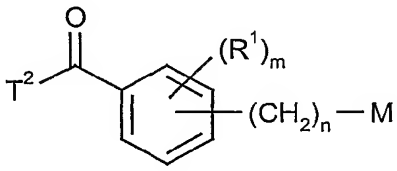


(VIII)

wherein

R^2 has the same definition as aforementioned

with compounds of the formula (IX)



(IX)

wherein

R^1, m and n have the same definition as aforementioned,

T^2 represents C_{1-4} alkyl, preferably methyl or ethyl, and

M represents halogen,

in an appropriate diluent, for example, N,N-dimethylformamide, in the presence of an appropriate condensing agent, for example, potassium carbonate.

The compounds of the above-mentioned formula (VIII) are known compounds described, for example, in Berichte Vol. 28, p. 74-76 (1895) and can be easily prepared according to the process described in said publication.

On the other hand, the compounds of the above-mentioned formula (IX), a part of which are novel compounds which were not described in the literature up to the

present, can be easily prepared according to the process described, for example, in Japanese Laid-open Patent Publication No. 173/1990.

The compounds of the formula (II), the starting materials in the above-mentioned preparation process (a), can also be easily prepared from compounds of the aforementioned formula (VI) according to the process described, for example, in WO93/18031.

As typical examples of the compounds of the formula (II) used as the starting materials in the aforementioned preparation process (a), the followings can be mentioned:

3-Oxo-1-cyclohexenyl 2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoate,
3-oxo-1-cyclohexenyl 2-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-4-fluorobenzoate,
3-oxo-1-cyclohexenyl 4-chloro-2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoate,
3-oxo-1-cyclohexenyl 4-chloro-2-{[(1-ethyl-1H-tetrazol-5-yl)thio]methyl}benzoate,
3-oxo-1-cyclohexenyl 4-chloro-2-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-benzoate,
3-oxo-1-cyclohexenyl 2-bromo-4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoate,
3-oxo-1-cyclohexenyl 4-bromo-2-{[(1-phenyl-1H-tetrazol-5-yl)thio]methyl}benzoate,
3-oxo-1-cyclohexenyl 2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-trifluoromethylbenzoate,
3-oxo-1-cyclohexenyl 2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-methylbenzoate,
3-oxo-1-cyclohexenyl 2,4-dichloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-benzoate,
3-oxo-1-cyclohexenyl 2,4-dichloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}benzoate,
3-oxo-1-cyclohexenyl 2,4-dichloro-3-{[(1-(2-chlorophenyl)-1H-tetrazol-5-yl)thio]-methyl}benzoate,
3-oxo-1-cyclohexenyl 2-chloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonylbenzoate,

3-oxo-1-cyclohexenyl 2-chloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonylbenzoate,

3-oxo-1-cyclohexenyl 2-chloro-3-{[(1-(n-pentyl)-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonylbenzoate,

3-oxo-1-cyclohexenyl 2-chloro-3-{[(1-(3-difluoromethylphenyl)-1H-tetrazol-5-yl)-thio]methyl}-4-methylsulfonylbenzoate,

3-oxo-1-cyclohexenyl 4-chloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-2-methylsulfanylbenzoate,

3-oxo-1-cyclohexenyl 2,4-dimethylsulfanyl-3-{[(1-methyl-1H-tetrazol-5-yl)thio]-methyl}benzoate,

3-oxo-1-cyclohexenyl 4-chloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-2-methylsulfonylbenzoate,

3-oxo-1-cyclohexenyl 2-chloro-4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoate,

3-oxo-1-cyclohexenyl 4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-2-methoxybenzoate,

3-oxo-1-cyclohexenyl 4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-2-methylsulfonyloxybenzoate,

3-oxo-1-cyclohexenyl 4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-2-nitrobenzoate,

3-oxo-1-cyclohexenyl 4-{[(1-ethyl-1H-tetrazol-5-yl)thio]methyl}-2-nitrobenzoate,

5,5-dimethyl-3-oxo-1-cyclohexenyl 2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-trifluoromethylbenzoate,

4,4-dimethyl-3-oxo-1-cyclohexenyl 2-bromo-4-{[(1-methyl-1H-tetrazol-5-yl)thio]-methyl}benzoate,

4,4-dimethyl-3-oxo-1-cyclohexenyl 2,4-dichloro-3-{[(1-methyl-1H-tetrazol-5-yl)-thio]methyl}benzoate,

4-{4-chloro-2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoyloxy}-bicyclo[3.2.1]-3-octen-2-one,

4-{2,4-dichloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-benzoyloxy}bicyclo[3.2.1]-3-octen-2-one,

4-{2-chloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonyl-benzoyloxy}bicyclo[3.2.1]-3-octen-2-one.

As typical examples of the compounds of the formula (IV) used as the starting materials in the preparation of the compounds of the aforementioned formula (II), the followings can be mentioned:

4-Chloro-2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoyl chloride,
4-bromo-2-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}benzoyl chloride,
2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-trifluoromethylbenzoyl chloride,
2,4-dichloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoyl chloride,
2,4-dichloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}benzoyl chloride,
2-chloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonyl-benzoyl chloride,
2-chloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonylbenzoyl chloride,
2-chloro-4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoyl chloride,
2-bromo-4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoyl chloride,
4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-2-nitro-benzoyl chloride,
2,4-dichloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoyl bromide,
2-chloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonyl-benzoyl bromide,
2-chloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonylbenzoyl bromide.

As typical examples of the compounds of the formula (VI) used as the starting materials in the preparation of the compounds of the aforementioned formula (IV), the followings can be mentioned:

4-Chloro-2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoic acid,
4-bromo-2-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}benzoic acid,
2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-trifluoromethylbenzoic acid,
2,4-dichloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoic acid,

2,4-dichloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}benzoic acid,
2-chloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonyl-benzoic acid,
2-chloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonylbenzoic acid,
2-chloro-4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoic acid,
2-bromo-4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoic acid,
4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-2-nitro-benzoic acid.

As typical examples of the compounds of the formula (VII) used as the starting materials in the preparation of the compounds of the aforementioned formula (VI), the followings can be mentioned.

Methyl 4-chloro-2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoate,
methyl 4-bromo-2-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-benzoate,
methyl 2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-trifluoromethyl-benzoate,
methyl 2,4-dichloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-benzoate,
methyl 2,4-dichloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-benzoate,
methyl 2-chloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonylbenzoate,
methyl 2-chloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonyl-benzoate,
methyl 2-chloro-4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoate,
methyl 2-bromo-4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoate,
methyl 4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-2-nitro-benzoate,
ethyl 2,4-dichloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoate,
ethyl 2-chloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-4-methylsulfonylbenzoate,
ethyl 2-chloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonyl-benzoate.

The compounds of the formula (Ib), starting materials in the aforementioned preparation process (b), are a part of the compounds of the formula (I) of the present invention and can be easily prepared according to the above-mentioned preparation

process (a).

As typical examples of the compounds of the formula (Ib) used as the starting materials in the aforementioned preparation process (b), the followings, included in the formula (I), can be mentioned:

2-{4-Chloro-2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoyl}-cyclohexane-1,3-dione,
2-{4-bromo-2-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}benzoyl}-cyclohexane-1,3-dione,
2-{2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-trifluoromethyl-benzoyl}cyclohexane-1,3-dione,
2-{2,4-dichloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoyl}-cyclohexane-1,3-dione,
2-{2,4-dichloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-benzoyl}cyclohexane-1,3-dione,
2-{2-chloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonylbenzoyl}-cyclohexane-1,3-dione,
2-{2-chloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonyl-benzoyl}cyclohexane-1,3-dione,
2-{2-chloro-4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoyl}-cyclohexane-1,3-dione,
2-{2-bromo-4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoyl}-cyclohexane-1,3-dione,
2-{4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-2-nitrobenzoyl}-cyclohexane-1,3-dione,
3-{2-chloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonyl-benzoyl}bicyclo[3.2.1]-octane-2,4-dione

As a halogenating agent used for the reaction with the compounds of the formula (Ib) in the preparation process (b) there can be mentioned, for example, thionyl chloride,

thionyl bromide, oxalyl dichloride, oxalyl dibromide etc.

The compounds of the formula (Ic), the starting materials in the aforementioned preparation process (c), are a part of the compounds of the formula (I) of the present invention and can be easily prepared according to the above-mentioned preparation process (b).

As typical examples of the compounds of the formula (Ic) used as the starting materials in the aforementioned preparation process (c), the followings, included in the formula (I), can be mentioned:

3-Chloro-2-{4-chloro-2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-benzoyl}-2-cyclohexen-1-one,

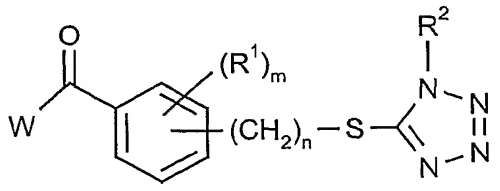
3-chloro-2-{4-bromo-2-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-benzoyl}-2-cyclohexen-1-one,
3-chloro-2-{2-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-trifluoromethylbenzoyl}-2-cyclohexen-1-one,
3-chloro-2-{2,4-dichloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-benzoyl}-2-cyclohexen-1-one,
3-chloro-2-{2,4-dichloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}benzoyl}-2-cyclohexen-1-one,
3-chloro-2-{2-chloro-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonyl-benzoyl}-2-cyclohexen-1-one,
3-chloro-2-{2-chloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonylbenzoyl}-2-cyclohexen-1-one,
3-chloro-2-{2-chloro-4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-benzoyl}-2-cyclo-hexen-1-one,
3-chloro-2-{2-bromo-4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-benzoyl}-2-cyclohexen-1-one,
3-chloro-2-{4-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}-2-nitrobenzoyl}-2-

cyclohexen-1-one,
4-chloro-2-{2-chloro-3-{[(1-cyclopropyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonylbenzoyl}bicyclo[3.2.1]-3-octen-2-one.

The compounds of the formula (III), the starting materials in the above-mentioned preparation process (c), are thiol compounds well known in the field of organic chemistry and as typical examples of the compounds of the formula (III) the followings can be mentioned:

Methyl mercaptan,
ethyl mercaptan,
thiophenol,
4-fluorothiophenol,
4-chlorothiophenol,
2-methylthiophenol,
4-ethylthiophenol,
4-trifluoromethylthiophenol etc.

Each compound of the formulae (II), (IV), (VI) and (VII), starting material or intermediate product in the aforementioned processes (a)-(c) for the preparation of the compounds of the formula (I) of the present invention is a novel compound which was not described in the literature up to the present. The compounds can be illustrated collectively by the following general formula (X)



(X)

wherein

W represents T^1, hydroxy or T^2, wherein

R^1, R^2, m, n, T^1, T^2 and M have the same definition as aforementioned.

The reaction of the aforementioned preparation process (a) can be conducted in an appropriate diluent. As examples of such diluents there can be mentioned aliphatic, alicyclic and aromatic hydrocarbons (which may optionally be chlorinated), for example, toluene, dichloromethane, chloroform and 1,2-dichloroethane; ethers, for example, ethyl ether, dimethoxyethane (DME) and tetrahydrofuran (THF); ketones, for example, methyl isobutyl ketone (MIBK); nitriles, for example, acetonitrile; esters, for example, ethyl acetate; acid amides, for example, dimethylformamide (DMF).

The preparation process (a) can be conducted in the presence of a cyanide and a base. As a cyanide usable in that case there can be mentioned, for example, sodium cyanide, potassium cyanide, acetone cyanohydrin and hydrogen cyanide. As a base there can be mentioned, for example, as inorganic bases, hydroxides and carbonates of alkali metals and alkaline earth metals, for example, sodium carbonate, potassium carbonate, lithium hydroxide, sodium hydroxide, potassium hydroxide and calcium hydroxide; and as organic bases, tertiary amines, dialkylaminoanilines and pyridines, for example, triethylamine, pyridine, 4-dimethylaminopyridine (DMAP), 1,4-diazabicyclo[2,2,2]octane (DABCO) and 1,8-diazabicyclo[5,4,0]undec-7-ene (DBU).

The aforementioned preparation process (a) can be conducted also in the co-existence of a phase-transfer catalyst. As examples of the phase-transfer catalyst usable in that case there can be mentioned crown ethers, for example, dibenzo-18-crown-6, 18-crown-6 and 15-crown-5.

The reaction of the preparation process (a) can be conducted in a substantially wide range of temperatures. Suitable temperatures are in the range of generally about -10 to about 80°C, preferably about 5 to about 40°C. Said reaction is conducted desirably under normal pressure. Optionally, however, it is possible to conduct it under elevated pressure or under reduced pressure.

In conducting the preparation process (a) the target compounds of the aforementioned formula (I), in case that Q represents groups (Q-1) or (Q-2), can be obtained, for example, by reacting 1 mole of a compound of the formula (II) with 1to 4 moles of triethylamine in a diluent, for example, acetonitrile, in the presence of 0.01 to 0.5 moles of acetone cyanohydrin.

In conducting the preparation process (a) it is possible to obtain the compounds of the formula (I) by conducting reactions starting from the compounds of the aforementioned formula (VI) continuously in one pot without isolating the compounds of the formulae (IV) and (II).

The reaction of the aforementioned preparation process (b) can be conducted in an appropriate diluent. As examples of such there can be mentioned aliphatic, alicyclic and aromatic hydrocarbons (which may optionally be chlorinated), for example, pentane, hexane, cyclohexane, petroleum ether, ligroine, benzene, toluene, xylene, dichloromethane, chloroform, carbon tetrachloride, 1,2-dichloroethane and chlorobenzene; ethers, for example, ethyl ether, methyl ethyl ether, isopropyl ether, butyl ether, dioxane, dimethoxyethane (DME), tetrahydrofuran (THF) and diethylene glycol dimethyl ether (DGM); ketones, for example, acetone, methyl ethyl ketone (MEK), methyl isopropyl ketone and methyl isobutyl ketone (MIBK); nitriles, for example, acetonitrile and propionitrile; esters, for example, ethyl acetate and amyl acetate; acid amides, for example, dimethylformamide (DMF), dimethylacetamide (DMA), N-methylpyrrolidone, 1,3-dimethyl-2-imidazolidinone and hexamethylphosphoric triamide (HMPA).

The reaction of the preparation process (b) can be conducted in a substantially wide range of temperatures. Suitable temperatures are in the range of generally about -20 to about 100°C, preferably about 0 to about 50°C. Said reaction is conducted desirably under normal pressure. Optionally, however, it is possible to conduct it under elevated pressure or under reduced pressure.

In conducting the preparation process (b) the target compounds of the aforementioned formula (I), in case that Q represents groups (Q-6) or (Q-7), wherein R^{11} in said group represents chloro or bromo, can be obtained, for example, by reacting 1 mole of a compound of the formula (Ib) with 1 to 5 moles of oxalyl dichloride in a diluent, for example, dichloromethane.

The reaction of the aforementioned preparation process (c) can be conducted in an appropriate diluent. As examples of such diluents there can be mentioned aliphatic, alicyclic and aromatic hydrocarbons (which may optionally be chlorinated), for example, pentane, hexane, cyclohexane, petroleum ether, ligroine, benzene, toluene, xylene, dichloromethane, chloroform, carbon tetrachloride, 1,2-dichloroethane, chlorobenzene and dichlorobenzene; ethers, for example, ethyl ether, methyl ethyl ether, isopropyl ether, butyl ether, dioxane, dimethoxyethane (DME), tetrahydrofuran (THF) and diethylene glycol dimethyl ether (DGM); ketones, for example, acetone, methyl ethyl ketone (MEK), methyl isopropyl ketone and methyl isobutyl ketone (MIBK); nitriles, for example, acetonitrile, propionitrile and acrylonitrile; esters, for example, ethyl acetate and amyl acetate; acid amides, for example, dimethylformamide (DMF), dimethylacetamide (DMA) and N-methylpyrrolidone; sulfones and sulfoxides, for example, dimethyl sulfoxide (DMSO) and sulfolane; bases, for example, pyridine.

The preparation process (c) can be conducted in the presence of a condensing agent. As a usable condensing agent there can be for example mentioned, as inorganic bases, hydrides and carbonates of alkali metals, for example, sodium hydride, lithium hydride, sodium carbonate and potassium carbonate; and as organic bases, tertiary amines, dialkylaminoanilines and pyridines, for example, triethylamine, 1,1,4,4-tetramethylethylenediamine (TMEDA), pyridine, 4-dimethylaminopyridine (DMAP), 1,4-diazabicyclo[2,2,2]octane (DABCO) and 1,8-diazabicyclo[5,4,0]undec-7-ene (DBU).

The reaction of the preparation process (c) can be conducted in a substantially wide

range of temperatures. Suitable temperatures are in the range of generally about -20 to about 140°C, preferably about 0 to about 100°C. Said reaction is conducted desirably under normal pressure. Optionally, however, it is possible to conduct it under elevated pressure or under reduced pressure.

In conducting the preparation process (c) the target compounds of the aforementioned formula (I), in case that Q represents groups (Q-3), (Q-4) or (Q-5) can be obtained, for example, by reacting 1 mole of a compound of the formula (Ic) with 1 to 5 moles of thiophenol in a diluent, for example, tetrahydrofuran in the presence of 1 to 5 moles of triethylamine.

The active compounds of the aforementioned formula (I), according to the present invention, show, as shown in the biological test examples to be described later, excellent herbicidal activities against various weeds and can be used as herbicides. In the present specification weeds mean, in the broadest sense, all plants which grow in locations where they are undesired. The compounds, acoording to the present invention, act as total or selective herbicides depending upon the applied concentration. The active compounds, according to the present invention, can be used , for example, between the following weeds and cultures.

Dicotyledon weeds of the genera: Sinapis, Lepidium, Galium, Stellaria, Chenopodium, Urtica, Senecio, Amaranthus, Portulaca, Xanthium, Ipomoea, Polygonum, Ambrosia, Cirsium, Sonchus, Solanum, Rorippa, Lamium, Veronica, Datura, Viola, Galeopsis, Papaver, Centaurea, Galinsoga, Rotala, Lindernia etc.

Dicotyledon cultures of the genera: Gossypium, Glycine, Beta, Daucus, Phaseolus, Pisum, Solanum, Linum, Ipomoea, Vicia, Nicotiana, Lycopersicon, Arachis, Brassica, Lactuca, Cucumis, Cucurbita etc.

Monocotyledon weeds of the genera: Echinochloa, Setaria, Panicum, Digitaria, Phleum, Poa, Festuca, Eleusine, Lolium, Bromus, Avena, Cyperus, Sorghum,

Agropyron, Monochoria, Fimbristylis, Sagittaria, Eleocharis, Scirpus, Paspalum, Ischaemum, Agrostis, Alopecurus, Cynodon etc.

Monocotyledon cultures of the genera: Oryza, Zea, Triticum, Hordeum, Avena, Secale, Sorghum, Panicum, Saccharum, Ananas, Asparagus, Allium etc.

The use of the compounds, according to the present invention, is not restricted to the above-mentioned plants, but may be applied to other plants in the same manner. The active compounds, according to the present invention, can, depending upon the applied concentration, non-selectively control weeds and can be used, for example, on industrial terrain, rail tracks, paths, places with or without tree plantings. Moreover, the active compounds, according to the present invention, can be used for controlling weeds in perennial cultures and applied in, for example, afforestations, decorative tree plantings, orchards, vineyards, citrus groves, nut orchards, banana plantations, coffee plantations, tea plantations, rubber plantations, oil palm plantations, cocoa plantations, soft fruit plantings, hopfields etc. and can be applied also for the selective controlling of weeds in annual cultures.

According to the invention all plants and plant parts can be treated. The term plants includes all plants and plant populations, such as desired or undesired wild plants and cultivated plants (including naturally occurring cultivated varieties). Cultivated plants can be plant varieties that were obtained by conventional breeding and optimizing processes or by biotechnological and genetic engineering methods or a combination of such processes and methods, including transgenic plants and including plant varieties that cannot or can be protected by plant patents or plant variety rights. Plant parts are all parts and organs of plants occurring above or below the surface of the soil, e.g. shoots, leaves, needles, stalks and stems, trunks, flowers, fruits and seeds as well as roots, tubers, bulbs and rhizomes. The term plant parts also includes harvested crops and propagation material, e.g. cuttings, tubers, bulbs, rhizomes, shoots and seeds.

According to the invention the plants and plant parts are treated using the usual methods by applying the active ingredients or compositions containing them directly to the plants or plant parts or to their surroundings (including the soil) or storeroom, e.g. by dipping, spraying, dusting, fogging, spreading and in the case of propagation material also by coating using one or multiple layers.

The active compounds, according to the present invention, can be made into the customary formulations. As such formulations there can be mentioned, for example, solutions, wettable powders, emulsions, suspensions, powders, water-dispersible granules, tablets, granules, suspension-emulsion concentrates, microcapsules in polymeric substances, jumbo formulations etc.

These formulations can be prepared according to per se known methods, for example, by mixing the active compounds with extenders, namely liquid or solid diluents or carriers, and optionally with surface-active agents, namely emulsifiers and/or dispersants and/or foam-forming agents.

As liquid diluents or carriers there can be mentioned, for example, aromatic hydrocarbons (for example, xylene, toluene, alkylnaphthalene etc.), chlorinated aromatic or chlorinated aliphatic hydrocarbons (for example, chlorobenzenes, ethylene chlorides, methylene chloride etc.), aliphatic hydrocarbons [for example, cyclohexane etc. or paraffins (for example, mineral oil fractions etc.)], alcohols (for example, butanol, glycol etc.) and their ethers, esters etc., ketones (for example, acetone, methyl ethyl ketone, methyl isobutyl ketone, cyclohexanone etc.), strongly polar solvents (for example, dimethylformamide, dimethyl sulphoxide etc.) and water. In case of using water as extender, for example, organic solvents can be used as auxiliary solvents.

As solid diluents or carriers there can be mentioned, for example, ground natural minerals (for example, kaolin, clay, talc, chalk, quartz, attapulgite, montmorillonite, diatomaceous earth etc.), ground synthetic minerals (for example, highly dispersed silicic acid, alumina, silicates etc.) etc. As solid carriers for granules there can be

mentioned, crushed and fractionated rocks (for example, calcite, marble, pumice, sepiolite, dolomite etc.), synthetic granules of inorganic and organic meals, particles of organic materials (for example, sawdust, coconut shells, maize cobs and tobacco stalks etc.) etc.

As emulsifiers and/or foam-forming agents there can be mentioned, for example, nonionic and anionic emulsifiers [for example, polyoxyethylene fatty acid esters, polyoxyethylene fatty acid alcohol ethers (for example, alkylaryl polyglycol ethers, alkylsulphonates, alkylsulphates, arylsulphonates etc.)], albumin hydrolysis products etc.

Dispersants include, for example, ligninsulphite waste liquor, methyl cellulose etc.

Tackifiers can also be used in formulations (powders, granules, emulsions). As said tackifiers there can be mentioned, for example, carboxymethyl cellulose, natural and synthetic polymers (for example, gum arabic, polyvinyl alcohol, polyvinyl acetate etc.).

Colorants can also be used. As said colorants there can be mentioned inorganic pigments (for example, iron oxide, titanium oxide, Prussian Blue etc.) and organic dyestuffs such as alizarin dyestuffs, azo dyestuffs or metal phthalocyanine dyestuffs, and further trace nutrients such as salts of metals such as iron, manganese, boron, copper, cobalt, molybdenum, zinc etc.

Said formulations can contain the active compounds of the formula (I) in a range of generally 0.1to 95 % by weight, preferably 0.5 to 90 % by weight.

The active compounds of the formula (I), according to the present invention, can be used as such or in their formulation forms for controlling weeds. They can be used also as a mixed agent with known herbicides. Such a mixed agent can be previously prepared as a final formulation form or can be prepared by tank-mixing on occasion

of application. As herbicides usable in combination with the compounds of the formula (I), according to the present invention, as a mixed agent there can be specifically mentioned, for example, the following herbicides shown in common names.

Acetamide type herbicides, for example, pretilachlor, butachlor, tenylchlor, alachlor etc.;
amide type herbicides, for example, clomeprop, etobenzanid etc.;
benzofuran type herbicides, for example, benfuresate etc.;
indanedione type herbicides, for example, indanofan etc.;
pyrazole type herbicides, for example, pyrazolate, benzofenap, pyrazoxyfen etc.;
oxazinone type herbicides, for example, oxaziclomefone etc.;
sulfonylurea type herbicides, for example, bensulfuron-methyl, azimsulfuron, imazosulfuron, pyrazosulfuron-ethyl, cyclosulfamron Ethoxysulfuron, Halosulfuron (-methyl) etc.;
thiocarbamate type herbicides, for example, thiobencarb, molinate, pyributycarb etc.;
triazine type herbicides, for example, dimethametryn Simetryn etc.;
triazole type herbicides, for example, cafenstrole etc.;
quinoline type herbicides, for example, quinclorac etc.;
isoxazole type herbicides, for example, isoxaflutole etc.;
dithiophosphate type herbicides, for example, anilofos etc.;
oxyacetamide type herbicides, for example, mefenacet, flufenacet etc.;
tetrazolinone type herbicides, for example, fentrazamide etc.;
dicarboxyimide type herbicides, for example, pentoxazone etc.;
trione type herbicides, for example, sulcotrione, benzobicyclon etc.;
phenoxypropinate type herbicides, for example, cyhalofop-butyl etc.;
benzoic acid type herbicides, for example, pyriminobac-methyl etc.;
diphenylether type herbicides, for example, chlomethoxyfen, oxyfluorfen etc.;
pyridinedicarbothioate type herbicides, for example, dithiopyr etc.;
phenoxy type herbicides, for example, MCPA, MCPB etc.;
urea type herbicides, for example, dymron, cumyluron etc.;

naphthalenedione type herbicides, for example, quinoclamine etc.;
isoxazolidinone type herbicides, for example, clomazone etc.
diphenylether type herbicides, for example, chlomethoxyfen, oxyfluorfen etc.;
pyridinedicarbothioate type herbicides, for example, dithiopyr etc.;
phenoxy type herbicides, for example, MCPA, MCPB etc.;
urea type herbicides, for example, dymron, cumyluron etc.;
naphthalenedione type herbicides, for example, quinoclamine etc.;
isoxazolidinone type herbicides, for example, clomazone etc.

In addition to the above mentioned herbicides, the following herbicides, shown in common names, for example, Acetochlor, Acifluorfen (-sodium), Aclonifen, Alloxydim (-sodium), Ametryne, Amicarbazone, Amidochlor, Amidosulfuron, Amitrole, Asulam, Atrazine, Azafenidin, Beflubutamid, Benazolin (-ethyl), Bentazon, Benzfendizone, Benzoylprop (-ethyl), Bialaphos, Bifenox, Bispyribac -(sodium), Bromacil, Bromobutide, Bromofenoxim, Bromoxynil, Butafenacil -(allyl), Butenachlor, Butralin, Butroxydim, Butylate, Carbetamide, Carfentrazone (-ethyl), Chloramben, Chloridazon, Chlorimuron (-ethyl), Chlornitrofen, Chlorsulfuron, Chlorthiamid, Chlortoluron, Cinidon (-ethyl), Cinmethylin, Cinosulfuron, Clefoxydim, Clethodim, Clodinafop (-propargyl), Clopyralid, Cloransulam (-methyl), Cyanazine, Cybutryne, Cycloate, Cycloxydim, 2,4-D, 2,4-DB, Desmedipham, Diallate, Dicamba, Dichlobenil, Dichlorprop (-P), Diclofop (-methyl), Diclosulam, Diethatyl (-ethyl), Difenopenten (-ethyl), Difenzoquat, Diflufenican, Diflufenzopyr, Dikegulac (-sodium), Dimefuron, Dimepiperate, Dimethachlor, Dimethenamid (-P), Dimexyflam, Dinitramine, Diphenamid, Diquat (-dibromide), Diuron, Epropodan, EPTC, Esprocarb, Ethalfluralin, Ethametsulfuron (-methyl), Ethiozin, Ethofumesate, Ethoxyfen, Fenoxaprop (-P-ethyl), Flamprop (-M-isopropyl, -M-methyl), Flazasulfuron, Florasulam, Fluazifop (-P-butyl), Fluazolate, Flucarbazone (-sodium), Fluchloralin, Flumetsulam, Flumiclorac (-pentyl), Flumioxazin, Flumipropyn, Fluometuron, Fluorochloridone, Fluoroglycofen (-ethyl), Flupoxam, Flupropacil, Flurpyrsulfuron (-methyl, -sodium), Flurenol (-butyl), Fluridone, Fluroxypyr (-butoxypropyl, -meptyl), Flurprimidol, Flurtamone,

Fluthiacet (-methyl), Fomesafen, Foramsulfuron, Glufosinate (-ammonium), Glyphosate (-ammonium, -isopropylammonium), Halosafen, Haloxyfop (-ethoxy-ethyl, -P-methyl), Hexazinone, Imazamethabenz (-methyl), Imazamethapyr, Imazamox, Imazapic, Imazapyr, Imazaquin, Imazethapyr, Iodosulfuron (-methyl, -sodium), Ioxynil, Isopropalin, Isoproturon, Isouron, Isoxaben, Isoxachlortole, Isoxadifen (-ethyl), Isoxapyrifop, Ketospiradox, Lactofen, Lenacil, Linuron, Mecoprop (-P), Mesotrione, Metamitron, Metazachlor, Methabenzthiazuron, Methyldymron, Metobenzuron, Metobromuron, (S-) Metolachlor, Metosulam, Metoxuron, Metribuzin, Metsulfuron (-methyl), Monolinuron, Naproanilide, Napropamide, Neburon, Nicosulfuron, Norflurazon, Orbencarb, Oryzalin, Oxadiargyl, Oxadiazon, Oxasulfuron, Paraquat, Pelargonsäure, Pendimethalin, Pendralin, Pethoxamid, Phenmedipham, Picolinafen, Piperophos, Primisulfuron (-methyl), Profluazol, Profoxydim, Prometryn, Propachlor, Propanil, Propaquizafop, Propisochlor, Propoxycarbazone (-sodium), Propyzamide, Prosulfocarb, Prosulfuron, Pyraflufen (-ethyl), Pyrazogyl, Pyribenzoxim, Pyridafol, Pyridate, Pyridatol, Pyriftalid, Pyrithiobac (-sodium), Quinmerac, Quizalofop (-P-ethyl, -P-tefuryl), Rimsulfuron, Sethoxydim, Simazine, Sulfentrazone, Sulfometuron (-methyl), Sulfosate, Sulfosulfuron, Tebutam, Tebuthiuron, Tepraloxydim, Terbuthylazine, Terbutryn, Thiazopyr, Thidiazimin, Thifensulfuron (-methyl), Tiocarbazil, Tralkoxydim, Triallate, Triasulfuron, Tribenuron (-methyl), Triclopyr, Tridiphane, Trifloxysulfuron, Trifluralin, Triflusulfuron (-methyl), Tritosulfuron.

The above-mentioned herbicides are known herbicides mentioned in "Pesticide Manual " 2000, published by The British Crop Protect Council.

The weight ratios of the groups of active substances in the mixed compositions can vary within relatively wide ranges.

For instance, per part by weight of (1) the compounds of the formula (I),

0.2 to 14 parts by weight of acetamide type herbicides,

preferably 0.66 to 5 parts by weight;
2 to 40 parts by weight of amide type herbicides,
preferably 3.96 to 16 parts by weight;
0.2 to 20 parts by weight of benzofuran type herbicides,
preferably 1.00 to 6 parts by weight;
0.2 to 8 parts by weight of indanedione type herbicides,
preferably 0.49 to 2 parts by weight;
0.06 to 4 parts by weight of oxazinone type herbicides,
preferably 0.20 to 0.8 parts by weight;
0.02 to 4 parts by weight of sulfonylurea type herbicides,
preferably 0.07 to 1.2 parts by weight;
1 to 100 parts by weight of thiocarbamate type herbicides,
preferably 2.47 to 40 parts by weight;
0.6 to 12 parts by weight of triazine type herbicides,
preferably 1.32 to 4.5 parts by weight;
0.1 to 8 parts by weight of triazole type herbicides,
preferably 0.33 to 3 parts by weight;
0.2 to 10 parts by weight of dithiophosphate type herbicides,
preferably 1.00 to 4 parts by weight;
0.2 to 50 parts by weight of oxyacetamide type herbicides,
preferably 1.00 to 12 parts by weight;
0.02 to 10 parts by weight of tetrazolinone type herbicides,
preferably 0.17 to 3 parts by weight;
0.1 to 12 parts by weight of dicarboxyimide type herbicides,
preferably 0.33 to 4.5 parts by weight;
0.2 to 12 parts by weight of phenoxypropinate type herbicides,
preferably 0.4 to 1.8 parts by weight;
0.6 to 20 parts by weight of diphenylether type herbicides,
preferably 1.65 to 7.5 parts by weight;
0.02 to 14 parts by weight of pyridinedicarbothioate type herbicides,
preferably 0.20 to 5 parts by weight;

0.2 to 10 parts by weight of phenoxy type herbicides,
preferably 0.66 to 4parts by weight,
and
2 to 80 parts by weight of urea type herbicides,
preferably 4.95 to 25 parts by weight,

are used.

Furthermore, the active compounds of the formula (I) , according to the present invention, can be mixed also with a safener and their application as a selective herbicide can be broadened to reduce phytotoxicity and to provide wider weed-control spectrum by such a mixing.

As an example of the safener, the following safeners can be mentioned;
AD-67, BAS-145138, Benoxacor, Cloquintocet (-mexyl), Cyometrinil, 2,4-D, DKA-24, Dichlormid, Dymron, Fenclorim, Fenchlorazol (-ethyl), Flurazole, Fluxofenim, Furilazole, Isoxadifen (-ethyl), MCPA, Mecoprop (-P), Mefenpyr (-diethyl), MG-191, Naphthalic anhydride, Oxabetrinil, PPG-1292, R-29148.

The above-mentioned safeners are known safeners mentioned in "Pesticide Manual", 2000, published by The British Crop Protect Council.

The weight ratios of the groups of active substances in the mixed comositions can vary within relatively wide ranges.

For instance, per part by weight of (1) the compounds of the formula (I),

0.05 to 50 parts by weight of Dichlormid,
preferably 0.1 to 10 parts by weight;
0.05 to 50 parts by weight of Dymron,
preferably 0.1 to 10 parts by weight;

0.05 to 50 parts by weight of Fenclorim,
preferably 0.1 to 10 parts by weight;
0.05 to 50 parts by weight of Mefenpyr (-diethyl),
preferably 0.1 to 10 parts by weight;
and
0.05 to 50 parts by weight of Naphthalic anhydride,
preferably 0.1 to 10 parts by weight,
are used.

And furthermore, the above-mentioned combinations of the compounds of the formula (I), according to the present invention, and the above-mentioned herbicides can be mixed with also the above-mentioned safeners and their application as selective herbicidal compositions can be broadened to reduce phytotoxicity and to provide wider weed - control spectrum by mixing safeners and/or other selective herbicides.

Surprisingly, some of the mixed compositions, according to the present invention show synergistic effects.

In case of using the active compounds of the formula (I) and their mixed compositions, according to the present invention, they can be directly used as such or used in formulation forms such as ready-to-use solutions, emulsions, tablets, suspensions, powders, pastes, granules or used in the use forms prepared by further dilution. The active compounds of the present invention can be applied by means of, for example, watering, spraying, atomizing, granule application etc.

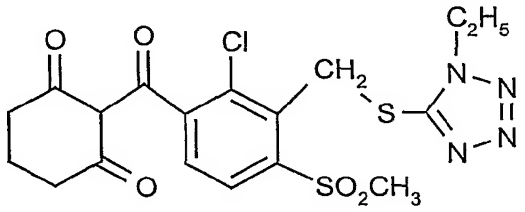
The active compounds of the formula (I) and their mixed compositions, according to the present invention, can be used at any stages before and after germination of plants. They can also be taken into the soil before sowing.

The application amount of the active compounds of the formula (I) and their mixed

compositions, according to the present invention, can be varied in a substantial range and are fundamentally different according to the nature of the desired effect. In case of using as herbicides, as the application amount there can be mentioned, for example, ranges of about 0.01 to about 3 kg, preferably about 0.05 to about 1 kg of the active compounds per hectare.

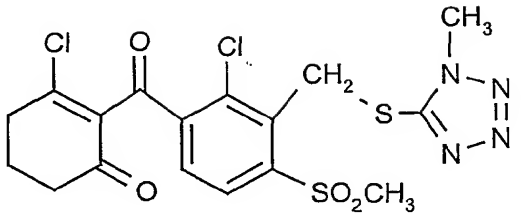
The preparations and applications of the compounds and their mixed compositions according to the present invention, will be described more specifically by the following examples. However, the present invention should not be restricted to them in any way.

Synthesis Example 1



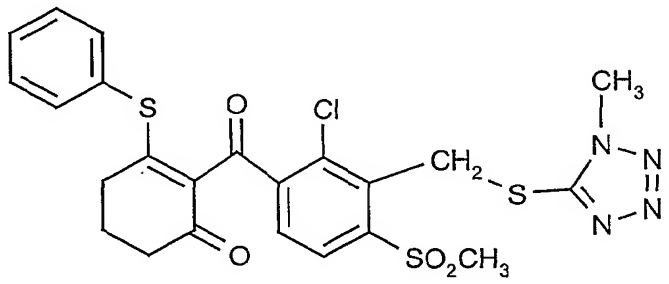
3-Oxo-1-cyclohexenyl 2-chloro-3-{[(1-ethyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonylbenzoate (0.83 g) was dissolved in acetonitrile (20 ml), to which triethylamine (0.34 g) and acetone cyanohydrin (10 mg) were added and the mixture was stirred at room temperature for 5 hours. After distilling off the solvent, the mixture was acidified by addition of diluted hydrochloric acid and extracted with dichloromethane (150 ml). The organic layer was washed with a saturated aqueous solution of sodium chloride and dried with anhydrous magnesium sulfate. Dichloromethane was distilled off to obtain the objected 2-{2-chloro-3-{[(1-ethyl-1H-tetrazol-1-yl)thio]methyl}-4-methylsulfonylbenzoyl}cyclohexane-1,3-dione (0.75 g). mp: 67-71°C.

Synthesis Example 2



To a solution of 2-{2-chloro-4-methylsulfonyl-3-{[(1-methyl-1H-tetrazol-5-yl)thio]-methyl}benzoyl}cyclohexane-1,3-dione (1.0 g) in dichloromethane (100 ml), oxalyl chloride (0.91 g) and 2 drops of N,N-dimethylformamide were added dropwise and the mixture was refluxed for 3 hours. The residue obtained by distilling off the solvent after the reaction was purified by silica gel column chromatography (eluant: ethyl acetate : hexane = 7 : 3) to obtain the objective 3-chloro-2-{2-chloro-4-methylsulfonyl-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoyl}-2-cyclohexen-1-one (0.71 g). IR (NaCl): .=1662, 1310, 1279, 1150 cm^{-1}.

Synthesis Example 3

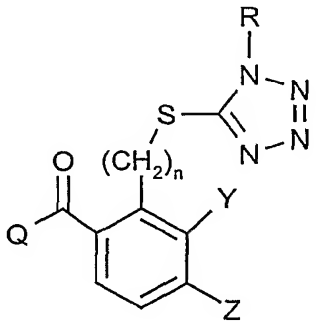


3-Chloro-2-{2-chloro-4-methylsulfonyl-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoyl}-2-cyclohexen-1-one (0.75 g) and thiophenol (0.19 g) were dissolved in tetrahydrofuran (7 ml), to which a solution of triethylamine (0.19 g) in tetrahydrofuran (3 ml) was added dropwise at 5°C and the mixture was stirred at room temperature for 4 hours. After the reaction cold water was added to the mixture, extracted with ethyl acetate (50 ml) and dried with anhydrous magnesium sulfate.

The residue obtained by distilling off the ethyl acetate was purified by silica gel column chromatography (eluant: ethyl acetate : hexane = 7 : 3) to obtain the objective 2-{2-chloro-4-methylsulfonyl-3-{[(1-methyl-1H-tetrazol-5-yl)thio]methyl}benzoyl}-3-phenylthio-2-cyclohexen-1-one (0.61 g). mp: 76-87°C.

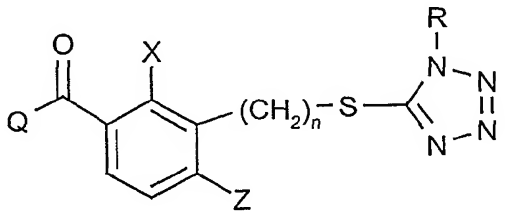
The compounds, obtained in the same manner as the above-mentioned Synthesis Examples 1-3, are shown in the following Tables 1-3, together with the compounds synthesized in the Synthesis Examples 1-3.

Examples of the compounds in case the compound of the formula (I) of the present invention is represented by the formula



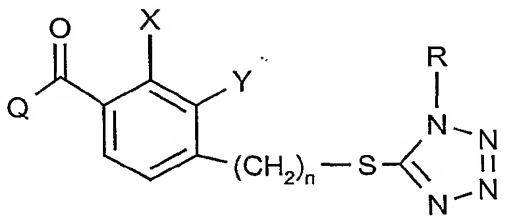
are shown in Table 1,

examples of the compounds in case they are represented by the following formula



are shown in Table 2, and

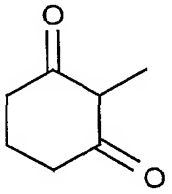
examples of the compounds in case they are represented by the following formula



are shown in Table 3.

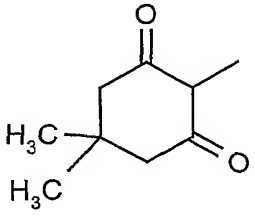
In Tables 1, 2 and 3,

Q1a a represents the group



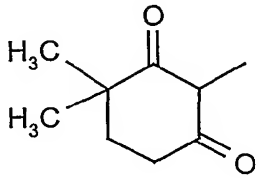
,

Q1b represents the group



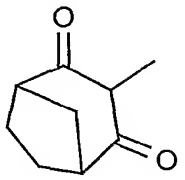
,

Q1c represents the group



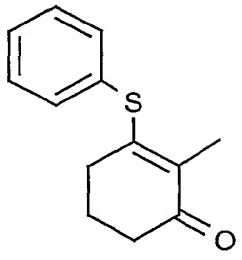
,

Q2 represents the group



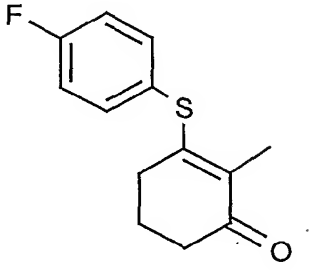
,

Q3a a represents the group



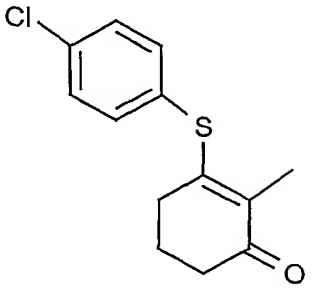
,

Q3b represents the group



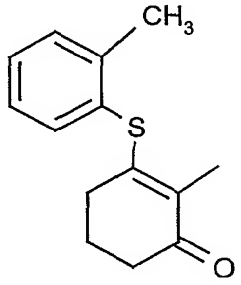
,

Q3c represents the group



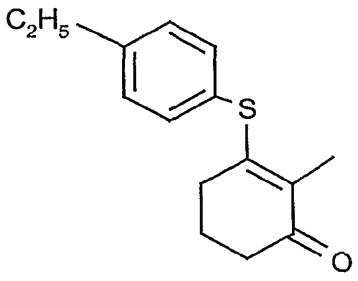
,

Q3d represents the group



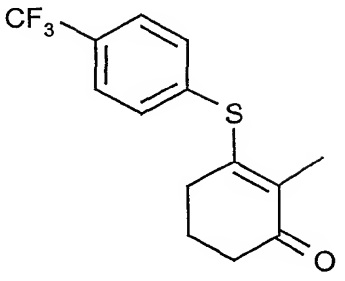
,

Q3e represents the group



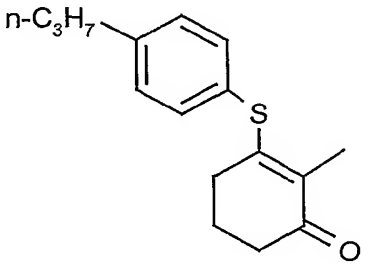
,

Q3f represents the group



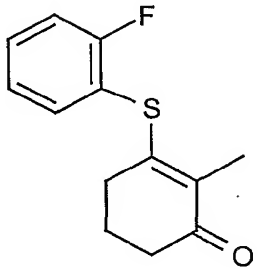
,

Q3g represents the group



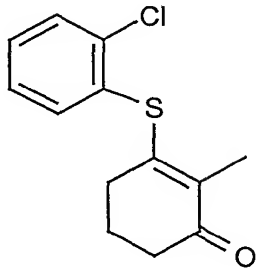
,

Q3h represents the group



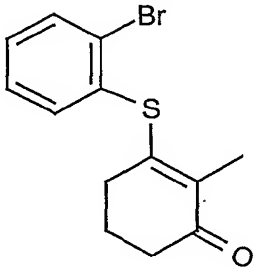
,

Q3i represents the group



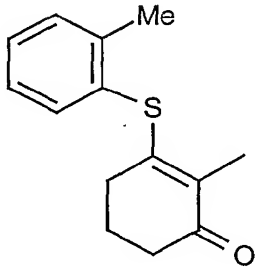
,

Q3j represents the group



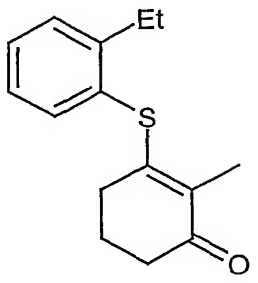
,

Q3k represents the group



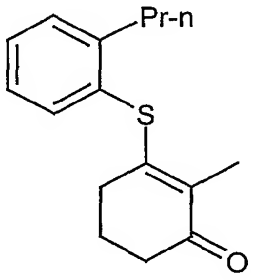
,

Q3l represents the group



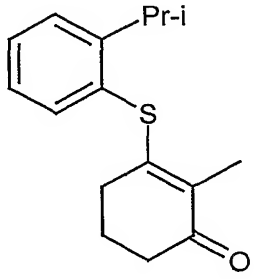
,

Q3m represents the group



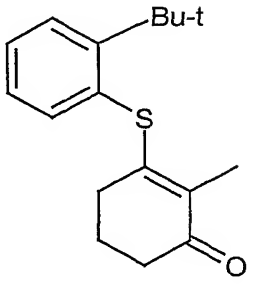
,

Q3n represents the group



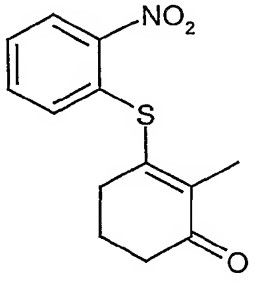
,

Q3o represents the group



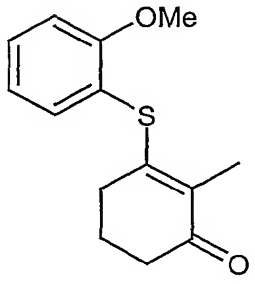
,

Q3p represents the group



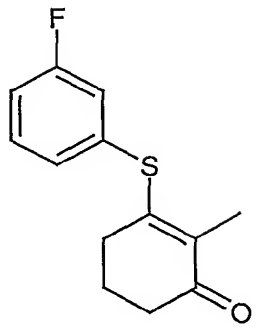
,

Q3q represents the group



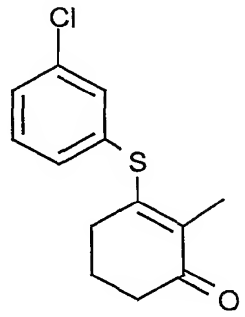
,

Q3r represents the group



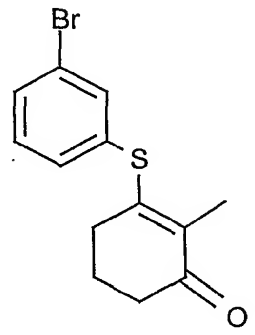
,

Q3s represents the group



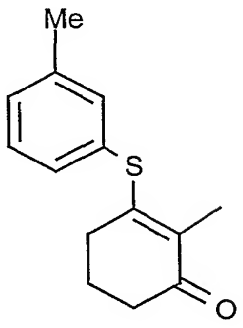
,

Q3t represents the group



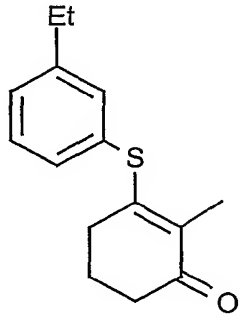
,

Q3u represents the group



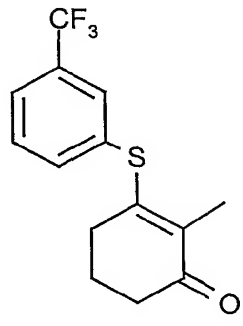
,

Q3v represents the group



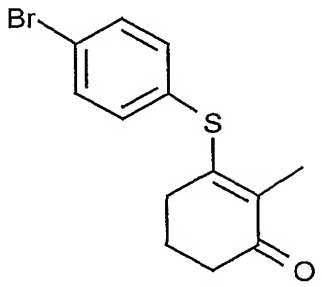
,

Q3w represents the group



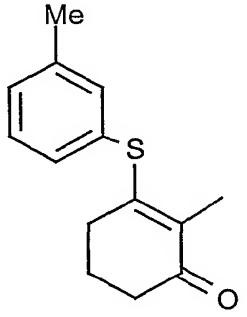
,

Q3x represents the group



,

Q3y represents the group



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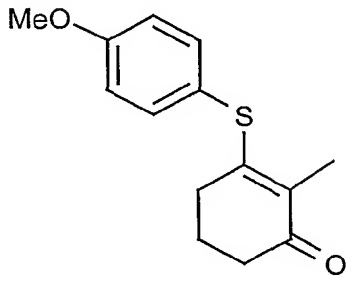
Q3z represents the group



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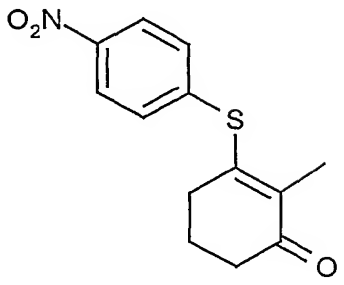
Q3za represents the group



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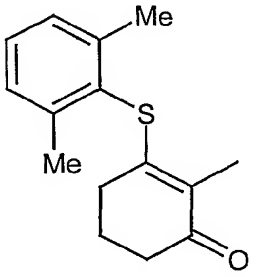
Q3zb represents the group



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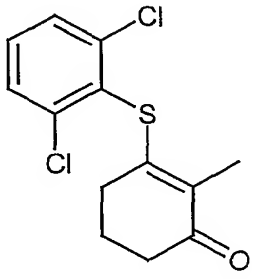

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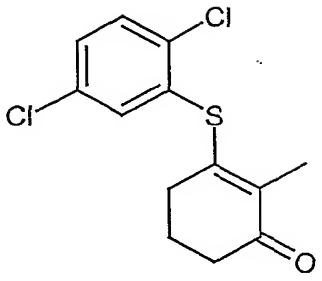
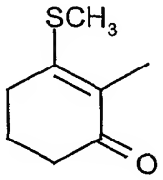


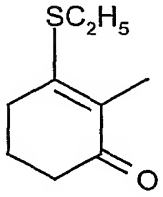
,

Q4a represents the group



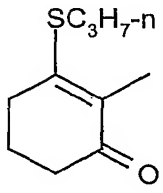
,

Q4b represents the group



,

Q4c represents the group



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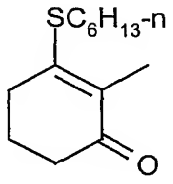
Q4d represents the group



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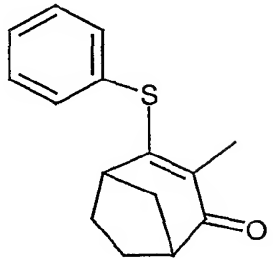
Q5a represents the group



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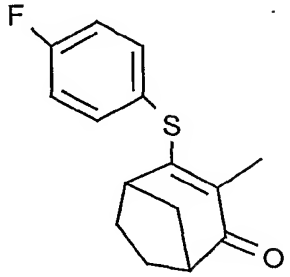
Q5b represents the group



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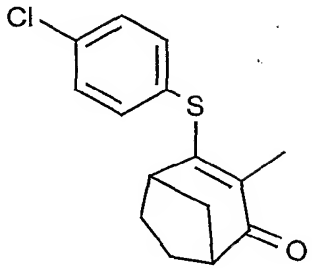
Q5c represents the group



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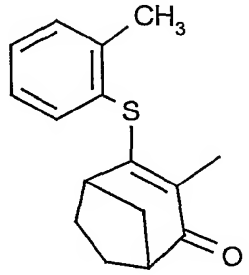
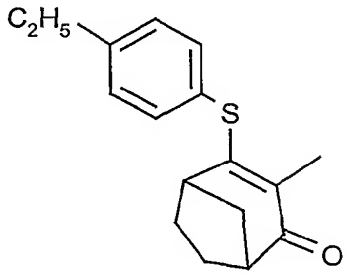
Q5d represents the group



,

Q5e represents the group



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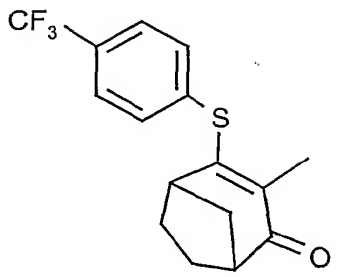
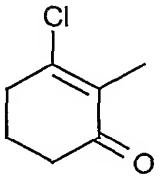
Q5f represents the group



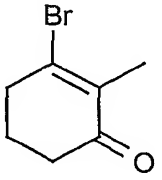
,

Q6a represents the group



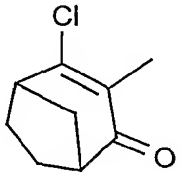
,

Q6b represents the group



,

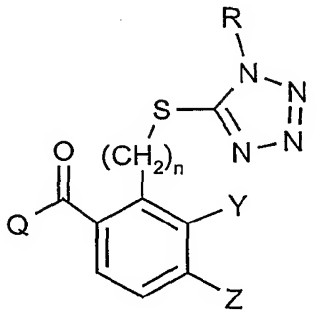
Q7 represents the group

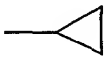
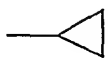


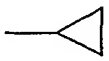
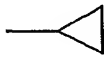
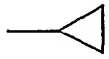
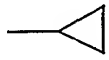
Me represents methyl, Et represents ethyl, n-Pr represents n-propyl, i-Pr represents isopropyl, n-Bu represents n-butyl, t-Bu represents tert-butyl,n-Hex represents n-

hexyl, OMe represents methoxy, OEt represents ethoxy, SMe represents methylthio, SEt represents ethylthio, SO_2Me represents methylsulfonyl, SO_2Et represents ethylsulfonyl, SO_2n-Pr represents n-propylsulfonyl, OSO_2Me represents methylsulfonyloxy, OSO_2Et represents ethylsulfonyloxy and Ph represents phenyl.

Table 1



Compound No.	Y	Z	R	n	Q	melting point (mp) or n_D^{20}
I-1	H	H	Me	1	Q1a	
I-2	H	H	Me	2	Q1a	
I-3	H	H	Me	1	Q2	
I-4	OMe	H	Me	1	Q1a	
I-5	Cl	H	Me	1	Q1a	
I-6	Me	H	Me	1	Q1a	
I-7	H	F	Me	1	Q1a	
I-8	H	F	Me	1	Q2	
I-9	H	F	Me	1	Q3a	
I-10	H	F	Me	1	Q5a	
I-11	H	F	Me	1	Q6a	
I-12	H	F	Et	1	Q1a	
I-13	H	F			Q1a	
I-14	H	Cl	Me	1	Q1a	
I-15	H	Cl	Me	2	Q1a	
I-16	H	Cl	Me	1	Q2	
I-17	H	Cl	Me	1	Q3a	
I-18	H	Cl	Me	1	Q5a	
I-19	H	Cl	Me	1	Q7	
I-20	H	Cl	Et	1	Q1a	
I-21	H	Cl		1	Q1a	
I-22	H	Br	Me	1	Q1a	1.6212
I-23	H	Br	Me	1	Q2	

Compound No.	Y	Z	R	n	Q	melting point (mp) or n_D^{20}
I-24	H	Br	Me	1	Q3a	
I-25	H	Br	Me	1	Q5a	
I-26	H	Br	Me	1	Q6a	
I-27	H	Br	Et	1	Q1a	
I-28	H	Br		1	Q1a	
I-29	H	I	Me	1	Q1a	
I-30	H	I	Me	1	Q2	
I-31	H	I	Me	1	Q3a	
I-32	H	I	Me	1	Q5a	
I-33	H	I	Et	1	Q1a	
I-34	H	I		1	Q1a	
I-35	H	Me	Me	1	Q1a	
I-36	H	CF_3	Me	1	Q1a	
I-37	H	CF_3	Me	1	Q2	
I-38	H	CF_3	Me	1	Q3a	
I-39	H	CF_3	Me	1	Q5a	
I-40	H	CF_3	Me	1	Q6a	
I-41	H	CF_3	Me	1	Q7	
I-42	H	CF_3	Et	1	Q1a	
I-43	H	CF_3		1	Q1a	
I-44	H	OMe	Me	1	Q1a	
I-45	H	OMe	Me	1	Q2	
I-46	H	OMe	Me	1	Q3a	
I-47	H	OMe	Me	1	Q5a	
I-48	H	OMe	Me	1	Q6a	
I-49	H	OMe	Et	1	Q1a	
I-50	H	OMe		1	Q1a	
I-51	H	OSO_2Me	Me	1	Q1a	
I-52	H	OSO_2Me	Me	1	Q2	
I-53	H	SMe	Me	1	Q1a	
I-54	H	SMe	Me	1	Q2	
I-55	H	SO_2Me	Me	1	Q1a	
I-56	H	SO_2Me	Me	1	Q2	
I-57	H	SO_2Me	Me	1	Q3a	
I-58	H	SO_2Me	Me	1	Q5	

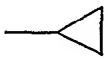
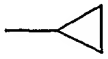
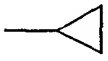
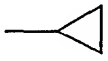
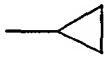
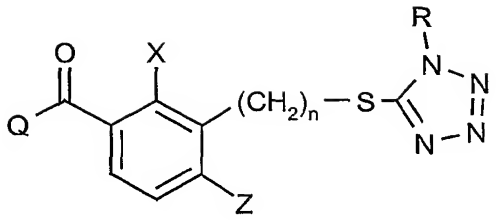
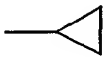
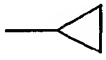
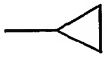
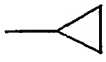
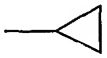
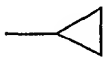
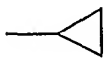
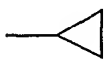
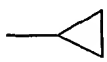
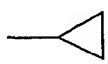
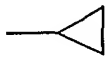
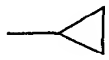
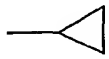
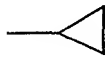
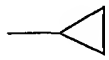
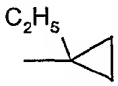
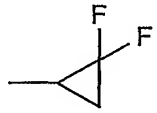
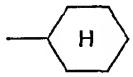
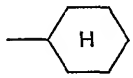
Compound No.	Y	Z	R	n	Q	melting point (mp) or n_D^{20}
I-59	H	SO_2Me		Me	1	Q6a
I-60	H	SO_2Me		Et	1	Q1a
I-61	H	SO_2Me			1	Q1a
I-62	H	NO_2		Me	1	Q1a
I-63	H	NO_2		Me	1	Q2
I-64	H	NO_2		Me	1	Q3a
I-65	H	NO_2		Me	1	Q5a
I-66	H	NO_2		Et	1	Q1a
I-67	H	NO_2			1	Q1a
I-68	H	CN		Me	1	Q1a
I-69	H	CN		Me	1	Q2
I-70	H	CN		Me	1	Q3a
I-71	H	CN		Me	1	Q5a
I-72	H	CN		Et	1	Q1a
I-73	H	CN			1	Q1a
I-74	H	$OCHF_2$		Me	1	Q1a
I-75	H	$OCHF_2$		Me	1	Q2
I-76	H	$OCHF_2$		Me	1	Q3a
I-77	H	$OCHF_2$		Me	1	Q5a
I-78	H	$OCHF_2$		Me	1	Q6a
I-79	H	$OCHF_2$		Et	1	Q1a
I-80	H	$OCHF_2$			1	Q1a
I-81	H	OCF_3		Me	1	Q1a
I-82	H	OCF_3		Me	1	Q2
I-83	H	OCF_3		Me	1	Q3a
I-84	H	OCF_3		Me	1	Q5a
I-85	H	OCF_3		Et	1	Q1a
I-86	H	OCF_3			1	Q1a

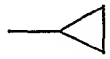
Table 2

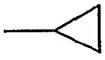
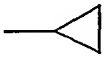
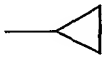
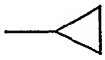
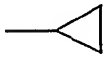
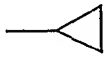


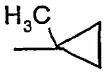
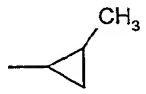
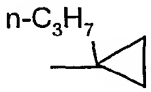
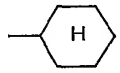
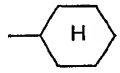
Compound No.	X	Z	R	n	Q	melting point (mp) or n_D^{20}
II-1	H	H	Me	1	Q1a	
II-2	H	H	Me	2	Q1a	
II-3	H	H	Me	1	Q2	
II-4	OMe	H	Me	1	Q1a	
II-5	OMe	H	Me	1	Q2	
II-6	OSO_2Me	H	Me	1	Q1a	
II-7	OSO_2Me	H	Me	1	Q2	
II-8	NO_2	H	Me	1	Q1a	
II-9	NO_2	H	Me	1	Q2	
II-10	F	Cl	Me	1	Q1a	66-72
II-11	F	Cl	Me	1	Q2	
II-12	F	Cl	Me	1	Q3a	
II-13	F	Cl	Me	1	Q5a	
II-14	F	Cl		1	Q1a	
II-15	F	Cl		1	Q2	
II-16	F	Cl		1	Q3a	
II-17	F	Cl		1	Q5a	
II-18	Cl	Cl	Me	1	Q1a	1.6010
II-19	Cl	Cl	Me	2	Q1a	
II-20	Cl	Cl	Me	1	Q1b	
II-21	Cl	Cl	Me	1	Q1c	
II-22	Cl	Cl	Me	1	Q2	
II-23	Cl	Cl	Me	1	Q3a	
II-24	Cl	Cl	Me	1	Q3b	
II-25	Cl	Cl	Me	1	Q3d	

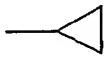
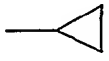
Compound No.	X	Z	R	n	Q	melting point (mp) or n_D^{20}
II-26	Cl	Cl	Me	1	Q4a	
II-27	Cl	Cl	Me	1	Q4b	
II-28	Cl	Cl	Me	1	Q5a	
II-29	Cl	Cl	Me	1	Q5c	
II-30	Cl	Cl	Me	1	Q6a	
II-31	Cl	Cl	Me	1	Q7	
II-32	Cl	Cl	Et	1	Q1a	
II-33	Cl	Cl	Et	2	Q1a	
II-34	Cl	Cl	Et	1	Q1b	
II-35	Cl	Cl	Et	1	Q1c	
II-36	Cl	Cl	Et	1	Q2	
II-37	Cl	Cl	Et	1	Q3a	
II-38	Cl	Cl	Et	1	Q3f	
II-39	Cl	Cl	Et	1	Q4a	
II-40	Cl	Cl	Et	1	Q4b	
II-41	Cl	Cl	Et	1	Q5a	
II-42	Cl	Cl	Et	1	Q5d	
II-43	Cl	Cl	Et	1	Q6a	
II-44	Cl	Cl	Et	1	Q7	
II-45	Cl	Cl	n-Pr	1	Q1a	
II-46	Cl	Cl	n-Pr	1	Q2	
II-47	Cl	Cl	n-Pr	1	Q3a	
II-48	Cl	Cl	n-Pr	1	Q5a	
II-49	Cl	Cl	i-Pr	1	Q1a	
II-50	Cl	Cl	i-Pr	1	Q2	
II-51	Cl	Cl	i-Pr	1	Q3a	
II-52	Cl	Cl	i-Pr	1	Q5a	
II-53	Cl	Cl		1	Q1a	
II-54	Cl	Cl		1	Q1b	
II-55	Cl	Cl		1	Q1c	
II-56	Cl	Cl		1	Q2	
II-57	Cl	Cl		1	Q3a	
II-58	Cl	Cl		1	Q3e	

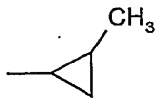
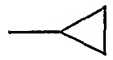
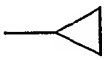
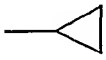
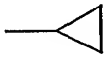
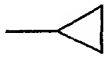
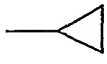
Compound No.	X	Z	R	n	Q	melting point (mp) or n_D^{20}
II-59	Cl	Cl		1	Q4a	
II-60	Cl	Cl		1	Q4b	
II-61	Cl	Cl		1	Q5a	
II-62	Cl	Cl		1	Q6a	
II-63	Cl	Cl		1	Q7	
II-64	Cl	Cl		1	Q1a	
II-65	Cl	Cl		1	Q1a	
II-66	Cl	Cl		1	Q1a	
II-67	Cl	Cl		1	Q2	
II-68	Cl	Cl	-CH=CH_2	1	Q1a	
II-69	Cl	Cl	-CH=CH_2	1	Q1b	
II-70	Cl	Cl	-CH=CH_2	1	Q1c	
II-71	Cl	Cl	-CH=CH_2	1	Q2	
II-72	Cl	Cl	-CH=CH_2	1	Q3a	
II-73	Cl	Cl	-CH=CH_2	1	Q4a	
II-74	Cl	Cl	-CH=CH_2	1	Q4b	
II-75	Cl	Cl	-CH=CH_2	1	Q5a	
II-76	Cl	Cl	-CH=CH_2	1	Q6a	
II-77	Cl	Cl	-CH_2CH=CH_2	1	Q1a	
II-78	Cl	Cl	-CH_2CH=CH_2	1	Q2	
II-79	Cl	Cl	-CH_2CH=CH_2	1	Q3a	
II-80	Cl	Cl	-CH_2CH=CH_2	1	Q5a	
II-81	Cl	Cl	Ph	1	Q1a	
II-82	Cl	Cl	2-Cl-Ph	1	Q1a	
II-83	Cl	Cl	2-Me-Ph	1	Q1a	
II-84	Cl	Cl	3-CF_3-Ph	1	Q1a	
II-85	Cl	Cl	CH_2CH_2F	1	Q1a	
II-86	Cl	Cl	CH_2CH_2F	1		

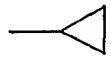
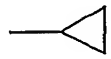
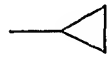
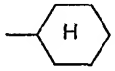
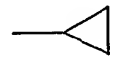
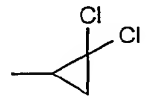
Compound No.	X	Z	R	n	Q	melting point (mp) or n_D^{20}
II-87	Cl	Cl	CH_2CH_2F	1	Q3a	
II-88	Cl	Cl	CH_2CH_2F	1	Q5a	
II-89	Cl	Cl	CH_2CH_2F	1	Q6a	
II-90	Cl	Cl	CH_2CH_2Cl	1	Q1a	
II-91	Cl	Cl	CH_2CH_2Cl	1	Q2	
II-92	Cl	Cl	CH_2CH_2Cl	1	Q3a	
II-93	Cl	Cl	CH_2CH_2Cl	1	Q5a	
II-94	Cl	Cl	CH_2CF_3	1	Q1a	
II-95	Cl	Cl	CH_2CF_3	1	Q1b	
II-96	Cl	Cl	CH_2CF_3	1	Q1c	
II-97	Cl	Cl	CH_2CF_3	1	Q2	
II-98	Cl	Cl	CH_2CF_3	1	Q3a	
II-99	Cl	Cl	CH_2CF_3	1	Q4a	
II-100	Cl	Cl	CH_2CF_3	1	Q4b	
II-101	Cl	Cl	CH_2CF_3	1	Q5a	
II-102	Cl	Cl	CH_2CF_3	1	Q6a	
II-103	Cl	Cl	CH_2CF_3	1	Q7	
II-104	Cl	Cl	$CH_2CF_2CF_3$	1	Q1a	
II-105	Cl	Cl	$CH_2CF_2CF_3$	1	Q2	
II-106	Cl	Cl	$CH_2CF_2CF_3$	1	Q3a	
II-107	Cl	Cl	$CH_2CH_2CH_2F$	1	Q5a	
II-108	Cl	Cl	$CH_2CH_2CH_2F$	1	Q1a	
II-109	Cl	Cl	$CH_2CH_2CH_2F$	1	Q2	
II-110	Cl	Cl	$CH_2CH_2CH_2F$	1	Q3a	
II-111	Cl	Cl	$CH_2CH_2CH_2F$	1	Q5a	
II-112	Cl	SMe	Me	1	Q1a	
II-113	Cl	SMe	Me	1	Q2	
II-114	Cl	SMe	Et	1	Q1a	
II-115	Cl	SMe		1	Q1a	
II-116	Cl	SMe	$-CH=CH_2$	1	Q1a	
II-117	Cl	SO_2Me	Me	1	Q1a	78-84
II-118	Cl	SO_2Me	Me	1	Q1a	
II-119	Cl	SO_2Me	Me	1	Q1b	
II-120	Cl	SO_2Me	Me	1	Q1c	
II-121	Cl	SO_2Me	Me	1	Q2	60-63
II-122	Cl	SO_2Me	Me	1	Q3a	76-87
II-123	Cl	SO_2Me	Me	1	Q3c	210-211
II-124	Cl	SO_2Me	Me	1	Q3	

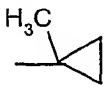
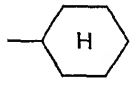
Compound No.	X	Z	R	n	Q	melting point (mp) or n_D^{20}
II-125	Cl	SO_2Me	Me	1	Q4a	79-82
II-126	Cl	SO_2Me	Me	1	Q4b	
II-127	Cl	SO_2Me	Me	1	Q5a	
II-128	Cl	SO_2Me	Me	1	Q5f	
II-129	Cl	SO_2Me	Me	1	Q6a	
II-130	Cl	SO_2Me	Me	1	Q7	
II-131	Cl	SO_2Me	Et	1	Q1a	67-71
II-132	Cl	SO_2Me	Et	2	Q1a	
II-133	Cl	SO_2Me	Et	1	Q1b	
II-134	Cl	SO_2Me	Et	1	Q1c	
II-135	Cl	SO_2Me	Et	1	Q2	
II-136	Cl	SO_2Me	Et	1	Q3a	
II-137	Cl	SO_2Me	Et	1	Q3b	
II-138	Cl	SO_2Me	Et	1	Q4a	
II-139	Cl	SO_2Me	Et	1	Q4b	
II-140	Cl	SO_2Me	Et	1	Q5a	
II-141	Cl	SO_2Me	Et	1	Q5b	
II-142	Cl	SO_2Me	Et	1	Q6a	
II-143	Cl	SO_2Me	Et	1	Q7	
II-144	Cl	SO_2Me	n-Pr	1	Q1a	142-145
II-145	Cl	SO_2Me	n-Pr	1	Q2	
II-146	Cl	SO_2Me	n-Pr	1	Q3a	
II-147	Cl	SO_2Me	n-Pr	1	Q5a	
II-148	Cl	SO_2Me	i-Pr	1	Q1a	69-73
II-149	Cl	SO_2Me	i-Pr	1	Q2	
II-150	Cl	SO_2Me	i-Pr	1	Q3a	
II-151	Cl	SO_2Me	i-Pr	1	Q5a	
II-152	Cl	SO_2Me		1	Q1a	79-84
II-153	Cl	SO_2Me		1	Q1b	
II-154	Cl	SO_2Me		1	Q1c	
II-155	Cl	SO_2Me		1	Q2	
II-156	Cl	SO_2Me		1	Q3a	
II-157	Cl	SO_2Me		1	Q3d	

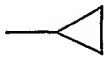
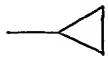
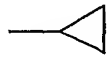
Compound No.	X	Z	R	n	Q	melting point (mp) or n_D^{20}
II-158	Cl	SO_2Me		1	Q4a	
II-159	Cl	SO_2Me		1	Q4b	
II-160	Cl	SO_2Me		1	Q5a	
II-161	Cl	SO_2Me		1	Q6a	
II-162	Cl	SO_2Me		1	Q7	
II-163	Cl	SO_2Me		1	Q1a	
II-164	Cl	SO_2Me		1	Q2	
II-165	Cl	SO_2Me		1	Q1a	
II-166	Cl	SO_2Me		1	Q1a	
II-167	Cl	SO_2Me		1	Q2	
II-168	Cl	SO_2Me	$-CH=CH_2$	1	Q1a	
II-169	Cl	SO_2Me	$-CH=CH_2$	1	Q1b	
II-170	Cl	SO_2Me	$-CH=CH_2$	1	Q1c	
II-171	Cl	SO_2Me	$-CH=CH_2$	1	Q2	
II-172	Cl	SO_2Me	$-CH=CH_2$	1	Q3a	
II-173	Cl	SO_2Me	$-CH=CH_2$	1	Q4a	
II-174	Cl	SO_2Me	$-CH=CH_2$	1	Q4b	
II-175	Cl	SO_2Me	$-CH=CH_2$	1	Q5a	
II-176	Cl	SO_2Me	$-CH=CH_2$	1	Q6a	
II-177	Cl	SO_2Me	$-CH_2CH=CH_2$	1	Q1a	63-68
II-178	Cl	SO_2Me	$-CH_2CH=CH_2$	1	Q2	
II-179	Cl	SO_2Me	$-CH_2CH=CH_2$	1	Q3a	
II-180	Cl	SO_2Me	$-CH_2CH=CH_2$	1	Q5a	
II-181	Cl	SO_2Me	Ph	1	Q1a	
II-182	Cl	SO_2Me	4-F-Ph	1	Q1a	
II-183	Cl	SO_2Me	2-Cl-Ph	1	Q1a	84-90
II-184	Cl	SO_2Me	3-Et-Ph	1	Q	

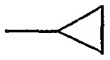
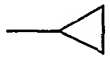
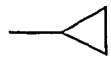
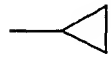
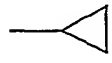
Compound No.	X	Z	R	n	Q	melting point (mp) or n_D^{20}
II-185	Cl	SO_2Me	4-NO_2-Ph	1	Q1a	
II-186	Cl	SO_2Me	CH_2CH_2F	1	Q1a	
II-187	Cl	SO_2Me	CH_2CH_2F	1	Q2	
II-188	Cl	SO_2Me	CH_2CH_2F	1	Q3a	
II-189	Cl	SO_2Me	CH_2CH_2F	1	Q5a	
II-190	Cl	SO_2Me	CH_2CH_2Cl	1	Q1a	
II-191	Cl	SO_2Me	CH_2CH_2Cl	1	Q2	
II-192	Cl	SO_2Me	CH_2CH_2Cl	1	Q3a	
II-193	Cl	SO_2Me	CH_2CH_2Cl	1	Q5a	
II-194	Cl	SO_2Me	CH_2CF_3	1	Q1a	82-87
II-195	Cl	SO_2Me	CH_2CF_3	1	Q1b	
II-196	Cl	SO_2Me	CH_2CF_3	1	Q1c	
II-197	Cl	SO_2Me	CH_2CF_3	1	Q2	
II-198	Cl	SO_2Me	CH_2CF_3	1	Q3a	
II-199	Cl	SO_2Me	CH_2CF_3	1	Q4a	
II-200	Cl	SO_2Me	CH_2CF_3	1	Q4b	
II-201	Cl	SO_2Me	CH_2CF_3	1	Q5a	
II-202	Cl	SO_2Me	CH_2CF_3	1	Q6a	
II-203	Cl	SO_2Me	CH_2CF_3	1	Q7	
II-204	Cl	SO_2Me	$CH_2CF_2CF_3$	1	Q1a	
II-205	Cl	SO_2Me	$CH_2CF_2CF_3$	1	Q2	
II-206	Cl	SO_2Me	$CH_2CF_2CF_3$	1	Q3a	
II-207	Cl	SO_2Me	$CH_2CH_2CH_2F$	1	Q5a	
II-208	Cl	SO_2Me	$CH_2CH_2CH_2F$	1	Q1a	
II-209	Cl	SO_2Me	$CH_2CH_2CH_2F$	1	Q2	
II-210	Cl	SO_2Me	$CH_2CH_2CH_2F$	1	Q3a	
II-211	Cl	SO_2Me	$CH_2CH_2CH_2F$	1	Q5a	
II-212	Cl	SO_2Et	Me	1	Q1a	70-74
II-213	Cl	SO_2Et	Me	1	Q2	
II-214	Cl	SO_2Et	Me	1	Q3a	
II-215	Cl	SO_2Et	Me	1	Q5a	
II-216	Cl	SO_2Et	Me	1	Q6a	
II-217	Cl	SO_2Et	Me	1	Q7	
II-218	Cl	SO_2Et	Et	1	Q1a	
II-219	Cl	SO_2Et	Et	1	Q2	
II-220	Cl	SO_2Et		1	Q1a	
II-221	Cl	SO_2Et		1	Q2	

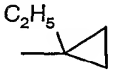
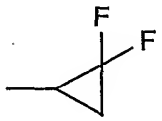
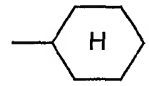
Compound No.	X	Z	R	n	Q	melting point (mp) or n_D^{20}
II-222	Cl	SO_2Et		1	Q1a	
II-223	Cl	SO_2Et	$-CH=CH_2$	1	Q1a	
II-224	Cl	SO_2Et	$-CH=CH_2$	1	Q2	
II-225	Cl	SO_2n-Pr		1	Q1a	
II-226	Br	Br	Me	1	Q1a	72-179
II-227	Br	Br	Me	1	Q1b	
II-228	Br	Br	Me	1	Q1c	
II-229	Br	Br	Me	1	Q2	
II-230	Br	Br	Me	1	Q3a	
II-231	Br	Br	Me	1	Q3c	
II-232	Br	Br	Me	1	Q3f	
II-233	Br	Br	Me	1	Q4a	
II-234	Br	Br	Me	1	Q4b	
II-235	Br	Br	Me	1	Q5a	
II-236	Br	Br	Me	1	Q5e	
II-237	Br	Br	Me	1	Q6a	
II-238	Br	Br	Me	1	Q7	
II-239	Br	Br	Et	1	Q1a	
II-240	Br	Br	Et	1	Q2	
II-241	Br	Br	Et	1	Q3a	
II-242	Br	Br	Et	1	Q3d	
II-243	Br	Br	Et	1	Q5a	
II-244	Br	Br	n-Pr	1	Q1a	
II-245	Br	Br	n-Pr	1	Q2	
II-246	Br	Br	i-Pr	1	Q1a	
II-247	Br	Br	i-Pr	1	Q2	
II-248	Br	Br		1	Q1a	
II-249	Br	Br		1	Q1b	
II-250	Br	Br		1	Q1c	
II-251	Br	Br		1	Q2	
II-252	Br	Br		1	Q3a	

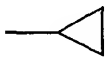
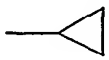
Compound No.	X	Z	R	n	Q	melting point (mp) or n_D^{20}
II-253	Br	Br		1	Q4a	
II-254	Br	Br		1	Q4b	
II-255	Br	Br		1	Q5a	
II-256	Br	Br		1	Q1a	
II-257	Br	Br	-CH=CH_2	1	Q1a	
II-258	Br	Br	-CH=CH_2	1	Q2	
II-259	Br	Br	-CH=CH_2	1	Q3a	
II-260	Br	Br	-CH=CH_2	1	Q5a	
II-261	Br	Br	-CH_2CH=CH_2	1	Q1a	
II-262	Br	Br	-CH_2CH=CH_2	1	Q2	
II-263	Br	Br	Ph	1	Q1a	
II-264	Br	Br	2-Cl-Ph	1	Q1a	
II-265	Br	Br	2-CF_3-Ph	1	Q1a	
II-266	Br	Br	CH_2CH_2F	1	Q1a	
II-267	Br	Br	CH_2CH_2F	1	Q2	
II-268	Br	Br	CH_2CH_2Cl	1	Q1a	
II-269	Br	Br	CH_2CH_2Cl	1	Q2	
II-270	Br	Br	CH_2CF_3	1	Q1a	
II-271	Br	Br	CH_2CF_3	1	Q2	
II-272	Br	Br	CH_2CF_3	1	Q3a	
II-273	Br	Br	CH_2CF_3	1	Q5a	
II-274	Br	Br	$CH_2CF_2CF_3$	1	Q1a	
II-275	Br	Br	$CH_2CH_2CH_2F$	1	Q1a	
II-276	Br	SO_2Me	Me	1	Q1a	87-90
II-277	Br	SO_2Me	Me	1	Q2	
II-278	Br	SO_2Me	Et	1	Q1a	
II-279	Br	SO_2Me		1	Q1a	
II-280	Br	SO_2Me		1	Q1a	
II-281	Br	SO_2Me	-CH=CH_2	1	Q1a	
II-282	OMe	Cl	Me	1	Q1a	1.6131
II-283	OMe	Cl	Me	1	Q1b	
II-284	OMe	Cl	Me	1	Q1c	

Compound No.	X	Z	R	n	Q	melting point (mp) or n_D^{20}
II-285	OMe	Cl	Me	1	Q2	
II-286	OMe	Cl	Me	1	Q3a	
II-287	OMe	Cl	Me	1	Q4a	
II-288	OMe	Cl	Me	1	Q4b	
II-289	OMe	Cl	Me	1	Q5a	
II-290	OMe	Cl	Me	1	Q6a	
II-291	OMe	Cl	Et	1	Q1a	
II-292	OMe	Cl	Et	1	Q2	
II-293	OMe	Cl	Et	1	Q3a	
II-294	OMe	Cl	Et	1	Q5a	
II-295	OMe	Cl	Et	1	Q7	
II-296	OMe	Cl	n-Pr	1	Q1a	
II-297	OMe	Cl	n-Pr	1	Q2	
II-298	OMe	Cl	i-Pr	1	Q1a	
II-299	OMe	Cl	i-Pr	1	Q2	
II-300	OMe	Cl		1	Q1a	
II-301	OMe	Cl		1	Q1b	
II-302	OMe	Cl		1	Q1c	
II-303	OMe	Cl		1	Q2	
II-304	OMe	Cl		1	Q3a	
II-305	OMe	Cl		1	Q4a	
II-306	OMe	Cl		1	Q4b	
II-307	OMe	Cl		1	Q5a	
II-308	OMe	Cl		1	Q1a	
II-309	OMe	Cl		1	Q1a	
II-310	OMe	Cl	-CH=CH$_2$	1	Q1a	
II-311	OMe	Cl	-CH=CH$_2$	1	Q2	
II-312	OMe	Cl	-CH=CH$_2$	1	Q3a	
II-313	OMe	Cl	-CH=CH$_2$	1	Q5a	

Compound No.	X	Z	R	n	Q	melting point (mp) or n_D^{20}
II-314	OMe	Cl	$-CH_2CH=CH_2$	1	Q1a	
II-315	OMe	Cl	$-CH_2CH=CH_2$	1	Q2	
II-316	OMe	Cl	Ph	1	Q1a	
II-317	OMe	Cl	2-Cl-Ph	1	Q1a	
II-318	OMe	Cl	CH_2CH_2F	1	Q1a	
II-319	OMe	Cl	CH_2CH_2F	1	Q2	
II-320	OMe	Cl	CH_2CH_2Cl	1	Q1a	
II-321	OMe	Cl	CH_2CH_2Cl	1	Q2	
II-322	OMe	Cl	CH_2CF_3	1	Q1a	
II-323	OMe	Cl	CH_2CF_3	1	Q2	
II-324	OMe	Cl	CH_2CF_3	1	Q3a	
II-325	OMe	Cl	CH_2CF_3	1	Q5a	
II-326	OMe	Cl	$CH_2CF_2CF_3$	1	Q1a	
II-327	OMe	Cl	$CH_2CH_2CH_2F$	1	Q1a	
II-328	$OCHF_2$	Cl	Me	1	Q1a	
II-329	$OCHF_2$	Cl	Me	1	Q2	
II-330	$OCHF_2$	Cl	Me	1	Q3a	
II-331	$OCHF_2$	Cl	Et	1	Q1a	
II-332	$OCHF_2$	Cl		1	Q1a	
II-333	$OCHF_2$	Cl	$-CH=CH_2$	1	Q1a	
II-334	OCH_2CF_3	Cl	Me	1	Q1a	
II-335	OCH_2CF_3	Cl	Me	1	Q2	
II-336	OCH_2CF_3	Cl	Et	1	Q1a	
II-337	SMe	Cl	Me	1	Q1a	
II-338	SMe	Cl	Me	1	Q2	
II-339	SMe	Cl	Et	1	Q1a	
II-340	SMe	Cl		1	Q1a	
II-341	SMe	Cl	$-CH=CH_2$	1	Q1a	
II-342	SMe	SMe	Me	1	Q1a	
II-343	SMe	SMe	Me	1	Q2	
II-344	SMe	SMe	Et	1	Q1a	
II-345	SMe	SMe		1	Q1a	
II-346	SMe	SMe	$-CH=CH_2$	1	Q1a	
II-347	SO_2Me	Cl	Me	1	Q1a	
II-348	SO_2Me	Cl	Me	1	Q2	
II-349	SO_2Me	Cl	Et	1	Q1a	

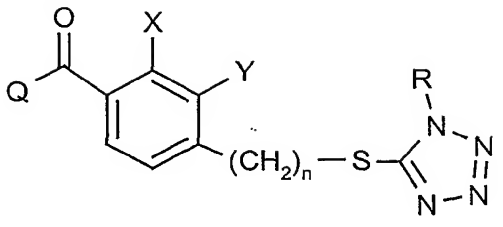
Compound No.	X	Z	R	n	Q	melting point (mp) or n_D^{20}
II-350	SO_2Me	Cl		1	Q1a	
II-351	SO_2Me	Cl	$-CH=CH_2$	1	Q1a	
II-352	SO_2Me	SO_2Me	Me	1	Q1a	
II-353	SO_2Me	SO_2Me	Me	1	Q2	
II-354	SO_2Me	SO_2Me	Et	1	Q1a	
II-355	SO_2Me	SO_2Me		1	Q1a	
II-356	SO_2Me	SO_2Me	$-CH=CH_2$	1	Q1a	
II-357	Me	SO_2Me	Me	1	Q1a	69-71
II-358	Me	SO_2Me	Me	2	Q1a	
II-359	Me	SO_2Me	Me	1	Q1b	
II-360	Me	SO_2Me	Me	1	Q1c	
II-361	Me	SO_2Me	Me	1	Q2	
II-362	Me	SO_2Me	Me	1	Q3a	
II-363	Me	SO_2Me	Me	1	Q3c	
II-364	Me	SO_2Me	Me	1	Q3d	
II-365	Me	SO_2Me	Me	1	Q4a	
II-366	Me	SO_2Me	Me	1	Q4b	
II-367	Me	SO_2Me	Me	1	Q5a	
II-368	Me	SO_2Me	Me	1	Q5c	
II-369	Me	SO_2Me	Me	1	Q6a	
II-370	Me	SO_2Me	Me	1	Q7	
II-371	Me	SO_2Me	Et	1	Q1a	
II-372	Me	SO_2Me	Et	1	Q2	
II-373	Me	SO_2Me	Et	1	Q3a	
II-374	Me	SO_2Me	Et	1	Q3b	
II-375	Me	SO_2Me	Et	1	Q5a	
II-376	Me	SO_2Me	n-Pr	1	Q1a	
II-377	Me	SO_2Me	n-Pr	1	Q2	
II-378	Me	SO_2Me	i-Pr	1	Q1a	
II-379	Me	SO_2Me	i-Pr	1	Q2	
II-380	Me	SO_2Me		1	Q1a	
II-381	Me	SO_2Me		1	Q1b	
II-382	Me	SO_2Me		1	Q1c	

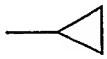
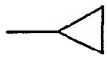
Compound No.	X	Z	R	n	Q	melting point (mp) or n_D^{20}
II-383	Me	SO_2Me		1	Q2	
II-384	Me	SO_2Me		1	Q3a	
II-385	Me	SO_2Me		1	Q4a	
II-386	Me	SO_2Me		1	Q4b	
II-387	Me	SO_2Me		1	Q5a	
II-388	Me	SO_2Me		1	Q1a	
II-389	Me	SO_2Me		1	Q1a	
II-390	Me	SO_2Me		1	Q1a	
II-391	Me	SO_2Me	$-CH=CH_2$	1	Q1a	
II-392	Me	SO_2Me	$-CH=CH_2$	1	Q2	
II-393	Me	SO_2Me	$-CH=CH_2$	1	Q3a	
II-394	Me	SO_2Me	$-CH=CH_2$	1	Q5a	
II-395	Me	SO_2Me	$-CH_2CH=CH_2$	1	Q1a	
II-396	Me	SO_2Me	$-CH_2CH=CH_2$	1	Q2	
II-397	Me	SO_2Me	Ph	1	Q1a	
II-398	Me	SO_2Me	2-Cl-Ph	1	Q1a	
II-399	Me	SO_2Me	4-NO_2-Ph	1	Q1a	
II-400	Me	SO_2Me	CH_2CH_2F	1	Q1a	
II-401	Me	SO_2Me	CH_2CH_2F	1	Q2	
II-402	Me	SO_2Me	CH_2CH_2Cl	1	Q1a	
II-403	Me	SO_2Me	CH_2CH_2Cl	1	Q2	
II-404	Me	SO_2Me	CH_2CF_3	1	Q1a	
II-405	Me	SO_2Me	CH_2CF_3	1	Q2	
II-406	Me	SO_2Me	CH_2CF_3	1	Q3a	
II-407	Me	SO_2Me	CH_2CF_3	1	Q5a	
II-408	Me	SO_2Me	$CH_2CF_2CF_3$	1	Q1a	
II-409	Me	SO_2Me	$CH_2CH_2CH_2F$	1	Q1a	
II-410	CN	SO_2Me	Me	1	Q1a	54-60
II-411	CN	SO_2Me	Me	1	Q2	
II-412	CN	SO_2Me	Me	1	Q3a	

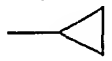
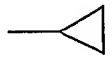
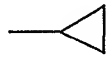
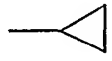
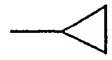
Compound No.	X	Z	R	n	Q	melting point (mp) or n_D^{20}
II-413	CN	SO_2Me	Et	1	Q1a	
II-414	CN	SO_2Me		1	Q1a	
II-415	CN	SO_2Me		1	Q5a	
II-416	Cl	SEt	Me	1	Q1a	
II-417	Cl	SO_2Me	Me	1	Q3g	
II-418	Cl	SO_2Me	Me	1	Q4c	
II-419	Cl	SO_2Me	Me	1	Q4d	
II-420	Cl	SO_2Me	Me	1	Q6b	
II-421	Cl	SO_2Me	n-Bu	1	Q1a	
II-422	Cl	SO_2Me	n-Hex	1	Q1a	
II-423	Cl	SO_2Me	$-CH_2CH=CHCH_3$	1	Q1a	
II-424	Cl	SO_2Me	$-(CH_2)_4CH=CH_2$	1	Q1a	
II-425	Cl	SO_2Me	4-(n-Pr)-Ph	1	Q1a	
II-426	Cl	SO_2Me	$4-(CH_2CH_2Cl)$-Ph	1	Q1a	
II-427	Cl	SO_2Me	$-(CH_2)_4Cl$	1	Q1a	
II-428	OEt	Cl	Me	1	Q1a	
II-429	OSO_2Me	Cl	Me	1	Q1a	
II-430	OSO_2Et	Cl	Me	1	Q1a	
II-431	Et	SO_2Me	Me	1	Q1a	
II-432	Cl	SO_2Me	Me	1	Q3h	
II-433	Cl	SO_2Me	Me	1	Q3i	128-131
II-434	Cl	SO_2Me	Me	1	Q3j	
II-435	Cl	SO_2Me	Me	1	Q3d	85-91
II-436	Cl	SO_2Me	Me	1	Q3k	
II-437	Cl	SO_2Me	Me	1	Q3l	
II-438	Cl	SO_2Me	Me	1	Q3m	
II-439	Cl	SO_2Me	Me	1	Q3n	
II-440	Cl	SO_2Me	Me	1	Q3o	
II-441	Cl	SO_2Me	Me	1	Q3p	
II-442	Cl	SO_2Me	Me	1	Q3q	
II-443	Cl	SO_2Me	Me	1	Q3r	

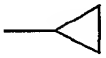
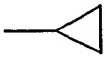
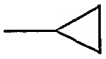
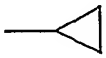
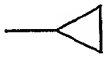
Com-pound No.	X	Z	R	n	Q	melting point (mp) or n_D^{20}
II-444	Cl	SO_2Me	Me	1	Q3s	
II-445	Cl	SO_2Me	Me	1	Q3t	
II-446	Cl	SO_2Me	Me	1	Q3u	
II-447	Cl	SO_2Me	Me	1	Q3v	
II-448	Cl	SO_2Me	Me	1	Q3w	
II-449	Cl	SO_2Me	Me	1	Q3b	
II-450	Cl	SO_2Me	Me	1	Q3x	
II-451	Cl	SO_2Me	Me	1	Q3y	208-209
II-452	Cl	SO_2Me	Me	1	Q3f	
II-453	Cl	SO_2Me	Me	1	Q3z	
II-454	Cl	SO_2Me	Me	1	Q3za	
II-455	Cl	SO_2Me	Me	1	Q3zb	
II-456	Cl	SO_2Me	Me	1	Q3zc	
II-457	Cl	SO_2Me	Me	1	Q3zd	

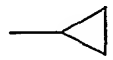
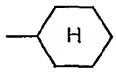
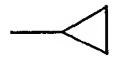
Table 3



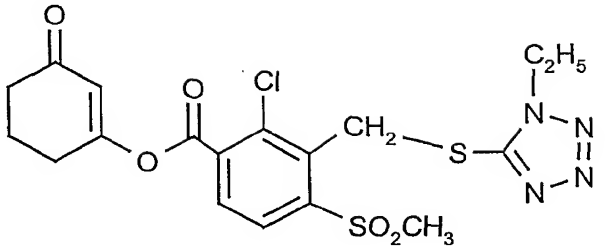
Compound No.	X	Y	R	n	Q	melting point (mp) or n_D^{20}
III-1	H	H	Me	1	Q1a	
III-2	H	H	Me	2	Q1a	
III-3	H	H	Me	1	Q2	
III-4	H	OMe	Me	1	Q1a	
III-5	H	NO_2	Me	1	Q1a	
III-6	F	H	Me	1	Q1a	
III-7	F	H	Me	1	Q2	
III-8	F	H	Et	1	Q1a	
III-9	F	H		1	Q1a	
III-10	F	H	-CH=CH_2	1	Q1a	
III-11	Cl	H	Me	1	Q1a	
III-12	Cl	H	Me	2	Q1a	
III-13	Cl	H	Me	1	Q2	
III-14	Cl	H	Me	1	Q3a	
III-15	Cl	H	Me	1	Q5a	
III-16	Cl	H	Me	1	Q6a	
III-17	Cl	H	Et	1	Q1a	
III-18	Cl	H		1	Q1a	
III-19	Cl	H	-CH=CH_2	1	Q1a	
III-20	Br	H	Me	1	Q1a	
III-21	Br	H	Me	1	Q2	
III-22	Br	H	Me	1	Q3a	
III-23	Br	H	Me	1	Q5a	
III-24	Br	H	Me	1	Q7	
III-25	Br	H	Et	1	Q1	

Compound No.	X	Y	R	n	Q	melting point (mp) or n_D^{20}
III-26	Br	H		1	Q1a	
III-27	Br	H	-CH=CH$_2$	1	Q1a	
III-28	I	H	Me	1	Q1a	
III-29	I	H	Me	1	Q2	
III-30	I	H	Me	1	Q3a	
III-31	I	H	Me	1	Q5a	
III-32	I	H	Me	1	Q6a	
III-33	I	H	Et	1	Q1a	
III-34	I	H		1	Q1a	
III-35	I	H	-CH=CH$_2$	1	Q1a	
III-36	CF_3	H	Me	1	Q1a	
III-37	CF_3	H	Me	1	Q2	
III-38	CF_3	H	Me	1	Q3a	
III-39	CF_3	H	Me	1	Q5a	
III-40	CF_3	H	Me	1	Q6a	
III-41	CF_3	H	Et	1	Q1a	
III-42	CF_3	H		1	Q1a	
III-43	CF_3	H	-CH=CH$_2$	1	Q1a	
III-44	OMe	H	Me	1	Q1a	
III-45	OMe	H	Me	1	Q2	
III-46	OMe	H	Me	1	Q3a	
III-47	OMe	H	Me	1	Q5a	
III-48	OMe	H	Et	1	Q1a	
III-49	OMe	H		1	Q1a	
III-50	OMe	H	-CH=CH$_2$	1	Q1a	
III-51	OSO_2Me	H	Me	1	Q1a	
III-52	OSO_2Me	H	Me	1	Q2	
III-53	OSO_2Me	H	Me	1	Q3a	
III-54	OSO_2Me	H	Me	1	Q5a	
III-55	OSO_2Me	H	Et	1	Q1a	
III-56	OSO_2Me	H		1	Q1a	
III-57	OSO_2Me	H	-CH=CH$_2$	1	Q1a	
III-58	SMe	H	Me	1	Q1a	
III-59	SMe	H	Me	1	Q	

Compound No.	X	Y	R	n	Q	melting point (mp) or n_D^{20}
III-60	SMe	H	Me	1	Q3a	
III-61	SMe	H	Et	1	Q1a	
III-62	SMe	H		1	Q1a	
III-63	SMe	H	-CH=CH$_2$	1	Q1a	
III-64	OSO_2Me	H	Me	1	Q1a	
III-65	OSO_2Me	H	Me	1	Q2	
III-66	OSO_2Me	H	Me	1	Q3a	
III-67	OSO_2Me	H	Me	1	Q5a	
III-68	OSO_2Me	H	Et	1	Q1a	
III-69	OSO_2Me	H		1	Q1a	
III-70	OSO_2Me	H	-CH=CH$_2$	1	Q1a	
III-71	NO_2	H	Me	1	Q1a	82-87
III-72	NO_2	H	Me	2	Q1a	
III-73	NO_2	H	Me	1	Q1b	
III-74	NO_2	H	Me	1	Q1c	
III-75	NO_2	H	Me	1	Q2	
III-76	NO_2	H	Me	1	Q3a	
III-77	NO_2	H	Me	1	Q4a	
III-78	NO_2	H	Me	1	Q4b	
III-79	NO_2	H	Me	1	Q5a	
III-80	NO_2	H	Me	1	Q6a	
III-81	NO_2	H	Me	1	Q7	
III-82	NO_2	H	Et	1	Q1a	
III-83	NO_2	H	Et	1	Q2	
III-84	NO_2	H	Et	1	Q3a	
III-85	NO_2	H	Et	1	Q5a	
III-86	NO_2	H	Et	1	Q6a	
III-87	NO_2	H	n-Pr	1	Q1a	
III-88	NO_2	H	n-Pr	1	Q2	
III-89	NO_2	H	i-Pr	1	Q1a	
III-90	NO_2	H	I-Pr	1	Q2	
III-91	NO_2	H		1	Q1a	
III-92	NO_2	H		1	Q2	
III-93	NO_2	H		1	Q3	

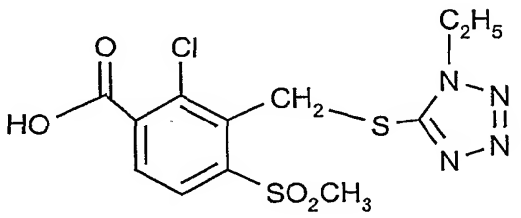
Compound No.	X	Y	R	n	Q	melting point (mp) or n_D^{20}
III-94	NO_2	H		1	Q5a	
III-95	NO_2	H		1	Q1a	
III-96	NO_2	H	$-CH=CH_2$	1	Q1a	
III-97	NO_2	H	$-CH=CH_2$	1	Q2	
III-98	NO_2	H	$-CH=CH_2$	1	Q3a	
III-99	NO_2	H	$-CH=CH_2$	1	Q5a	
III-100	NO_2	H	$-CH_2CH=CH_2$	1	Q1a	
III-101	NO_2	H	Ph	1	Q1a	
III-102	NO_2	H	2-Cl-Ph	1	Q1a	
III-103	NO_2	H	CH_2CH_2F	1	Q1a	
III-104	NO_2	H	CH_2CH_2Cl	1	Q1a	
III-105	NO_2	H	CH_2CF_3	1	Q1a	
III-106	NO_2	H	CH_2CH_2F	1	Q1a	
III-107	NO_2	H	$CH_2CF_2CF_3$	1	Q1a	
III-108	NO_2	H	$CH_2CH_2CH_2F$	1	Q1a	
III-109	CN	H	Me	1	Q1a	
III-110	CN	H	Me	1	Q2	
III-111	CN	H	Me	1	Q3a	
III-112	CN	H	Me	1	Q5a	
III-113	CN	H	Et	1	Q1a	
III-114	CN	H		1	Q1a	
III-115	CN	H	$-CH=CH_2$	1	Q1a	

Synthesis Example 4



2-Chloro-3-{[(1-ethyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonyl-benzoic acid (0.77 g) and thionyl chloride (0.49 g) were added to 1,2-dichloroethane (30 ml) and the mixture was, after addition of 2 drops of N,N-dimethylformamide, refluxed for 3 hours. After cooling, the residue obtained by distilling off the solvent was dissolved in dichloromethane (10 ml) and the mixture was added dropwise to a solution of 1,3-cyclohexanedione (0.28 g) and triethylamine (0.28 g) in dichloromethane (10 ml) at 5°C and stirred at room temperature for 6 hours. After the reaction the mixture was extracted with dichloromethane (100 ml), washed with diluted hydrochloric acid and an aqueous dolution of sodium hydrogen carbonate, and dried with anhydrous magnesium sulfate. The residue obtained by distilling off the dichloromethane was purified by silica gel column chromatography (eluant: ethyl acetate : hexane = 3 : 7) to obtain the objective 3-oxo-1-cyclohexenyl 2-chloro-3-{[(1-ethyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonylbenzoate (0.83 g). mp: 122-123°C.

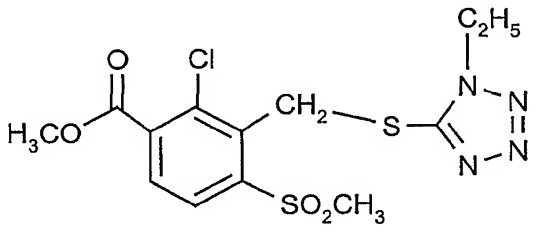
Synthesis Example 5



To a solution of methyl 2-chloro-3-{[(1-ethyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonylbenzoate (0.83 g) in dioxane (15 ml), a 10N aqueous solution of sodium

hydroxide (1.0 ml) and water (2 ml) were added and the mixture was stirred at room temperature for 3 hours. Water (30 ml) is added. Then, after concentration under reduced pressure, a 10N aqueous solution of sodium hydroxide (1.0 ml) was added to the concentrate and the concentrate is washed with ethyl acetate (100 ml). The aqueous layer was acidified with hydrochloric acid and extracted with ethyl acetate. The organic layer was washed with saturated aqueous solution of sodium chloride and dried with anhydrous magnesium sulfate. Ethyl acetate was distilled off to obtain the objected 2-chloro-3-{[(1-ethyl-1H-tetrazol-5-yl)thio]methyl}-4-methyl-sulfonylbenzoic acid (0.80 g). mp: 193-195°C.

Synthesis Example 6



1-Ethyl-5-mercaptotetrazole (0.31 g) and methyl 3-bromomethyl-2-chloro-4-methyl-sulfonylbenzoate (0.80 g) were suspended in acetonitrile (20 ml) and the suspension was, after addition of potassium carbonate (0.32 g), refluxed for 3 hours. After addition of cold water upon the completion of the reaction, the mixture was extracted with ethyl acetate (100 ml) and dried with anhydrous magnesium sulfate. The residue obtained by distilling off the ethyl acetate was recrystallized from dichloromethane-hexane to obtain the objected methyl 2-chloro-3-{[(1-ethyl-1H-tetrazol-5-yl)thio]methyl}-4-methylsulfonyl-benzoate (0.88 g). mp: 109-110°C.

Test Example 1: Test for herbicidal effect against paddy field weeds

Preparation of a formulation of the active compound

Carrier: Acetone 5 parts by weight
Emulsifier: Benzyloxypolyglycolether 1 part by weight

A formulation of the active substance is obtained as an emulsion by mixing 1 part by weight of the active compound with the above-mentioned amount of carrier and emulsifier. A prescribed amount of the formulation is diluted with water.

Test method

In a greenhouse 3 seedlings of paddy rice (cultivar: Nipponbare) of 2.5 leafstage (15 cm tall) were planted in a 500 cm^2 pot filled with paddy field soil. Then seeds or tubers of smallflower, bulrush, monochoria, broad-leaved weeds (common false pimpernel, Indian toothcup, long stemmed water wort, *Dopatrium junceum* Hammilt etc.) and Japanese ribbon wapato were inoculated and water was poured to a depth of about 2-3 cm.

5 Days after the rice transplantation a formulation of each active compound prepared according to the aforementioned preparation method was applied to the surface of the water. The herbicidal effect was examined after 3 weeks from the treatment during which period the water depth of 3 cm was maintained. The herbicidal effect was rated as 100% in the case of complete extiinction and as 0% in the case of no herbicidal effect.

As a result, the compounds No. II-18, II-117, II-122, II-131, II-194, II-212 and III-71 showed at the application rate of 0.25 kg/ha a herbicidal effect of more than 90% against paddy field weeds and showed safety to the transplanted paddy rice.

Test Example 2: Test of pre-emergence soil treatment against field weeds

Test method

In a greenhouse, on the surface layer of a 120 cm^2 pot filled with field soil, and then seeds of barnyardgrass, foxtail, common amaranth and knotweed were sown and covered with soil. The prescribed amount of chemicals prepared in the same manner as in the above-mentioned Test Example 1 was spread uniformly on the soil surface layer of each test pot. The herbicidal effect was examined after 4 weeks from the treatment.

Effects:
The compounds No. II-117, II-122 and II-194 showed at application rate of 2.0 kg/ha herbicidal activities of more than 90% against objective weeds (barnyardgrass, foxtail, common amaranth and knotweed).

Test Example 3: Test of post-emergence foliage treatment against field weeds

Test method

In a greenhouse, seeds of barnyardgrass, foxtail, common amaranth and knotweed were sown in 120 cm^2 pots filled with field soil and covered with soil. After 10 days after the sowing and soil covering (weeds were 2-leafstage in average) the prescribed amount of chemicals prepared in the same manner as in the above-mentioned Test Example 1 was spread uniformly on the foliage of the test plants in each test pot. The herbicidal effect was examined after 3 weeks from the treatment.

Results:

The compounds No. II-18, II-117, II-122, II-131, II-194, II-212 and II-276 showed at the chemical amount of 2.0 kg/ha herbicidal activities of more than 90% against

barnyardgrass, foxtail, common amaranth and knotweed.

Test Example 4: Test for synergistic action by foliar spray application

Preparation of the test solution

Carrier: acetone, 5 parts by weight
Emulsifier: benzyloxypolyglycol ether, 1 parts by weight

One part of an active compound and the above amounts of carrier and emulsifier are mixed to obtain a formulation of the active substance as an emulsion. A prescribed amount of this formulation is diluted with water to prepare testing solutions.

Test method

In a greenhouse, paddy soil was filled in pots (250 cm^2), and seeds of weed (barnyardgrass, bulrush, monochoria and falsepimpernel) were inoculated in the surface layer of the soil in the pots under wet conditions and covered with soil. All of the weed species were individually inoculated in each pot. Each pot was watered to 2 cm in depth. When the weeds grew up to 1.5-2.2 leaf stage (or pair), a predetermined amount of the compound as a testing solution prepared in the above was applied to the weeds in pots by foliar spray after draining the water in the pot. On the day following the application, the pots were irrigated again to 2 cm of water depth. The herbicidal effect was evaluated at 4 weeks after the application on a scale of 0(not active) to 100(complete damage).

Test results of test example 4 are shown in Table 4.

Test Example 5: Test for synergistic action by water sureface application

Test method

In a greenhouse, paddy soil was filled in pots (250 cm^2), and seeds of weed (barnyardgrass, bulrush, monochoria, falsepimpernel, indian toothcup, waterwort and flatstage) were inoculated in the surface layer of the soil in the pots under wet conditions and covered with soil. All of the weed species were individually inoculated in each pot. Each pot was watered to 2 cm in depth and the depth was kept during the test period. When the weeds grew up to 1.5-2.2 leaf stage (or pair), a predetermined amount of the compound as a testing solution prepared in the same manner as the above-mentioned Test Example 4 was applied to the pots by water surface treatment method. The herbicidal effect was evaluated at 4 weeks after the application on the same scale as in thetest method of Test Example 4.

Test results of test example 5 are shown in Table 5.

Synergistic action of Test Example 4 and Test Example 5 were evaluated by Colby's equation.

$$\text{Colby : } E = X + \left[Y \times \frac{(100-X)}{100} \right]$$

E : expected herbicidal activity at p+q g/ha

X : the percentage of herbicidal activity at p g/ha

Y : the percentage of herbicidal activity at q g/ha

The following abbreviations are used in Table 4 and Table 5 :

CYPSE	represents Cyperus serotinus (flatstage),
ECHSS	represents Echinochloa spp. (barnyardgrass),
ELTTP	reprerstnts Elatine triandra (waterwort),
LIDPY	represents Lindernia pyridaria (flasepimpernel),

MOOVP represents Monochoria vaginalis (monochoria),
ROTIN represents Rotala indica (indian toothcup),

Compounds (1) in Table 4 and Table 5 are listed by the compound numbers previously used in Tables 1, 2 and 3.

In Table 4 and Table 5 other known herbicides are represented by the capital letters as shown in the following list:

A: 4-(2-chlorophenyl)-N-cyclohexyl-N-ethyl-4,5-dihydro-5-oxo-1H-tetrazole –1-carboxamide (fentrazamide),

B: 3,'4'-dichloropropionanilide (propanil),

C: N,N-diethyl-3-mesitylsulfonyl-1H-1,2,4-triazole-1-carboxamide (cafenstrole),

D: 3-[1-(3,5-dichlorophenyl)-1-methylethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one (oxaziclomefone),

E: 2-chloro-2',6'-diethyl-N-(2-propoxyethyl) acetamide (pretilachlor),

F: 2-(1,3-benzothiazol-2-yloxy)-N-methylacetanilide (mefenacet),

G: (RS)-2-[2-(3-chlorophenyl)2,3-epoxypropyl]-2-ethylindan-1,3-dione (indanofan).

Table 4: Herbicidal efficacy (%) by foliar spray application

Test plant	Herbicidal efficacy (%)			Expected activity E according to Colby (%)
	compound(1) (g a.i./ha)	known herbicide (g a.i./ha)	compound(1) + known herbicide (g a.i./ha)	
1st run:	II-131 (125)	A (135)	II-131 + A (125 + 135)	II-131 + A
SCPSS	70	60	90	88
MOOVP	70	60	95	88
LIDPY	50	50	80	75
2nd run:	II-131 (125)	B (750)	II-131 + B (125 + 750)	II-131 + B
ECHSS	40	30	80	58
SCPSS	70	10	80	73
MOOVP	70	30	90	79
LIDPY	50	40	80	70

Table 5: Herbicidal efficacy (%) by water surface application

Test plant	Herbicidal efficacy (%)			Expected activity E According to Colby (%)
	compound(1) (g a.i./ha)	known herbicide (g a.i./ha)	compound(1) + known herbicide (g a.i./ha)	
1st run:	II-117 (75)	A (100)	II-117 + A (75 + 100)	II-117 + A
LIDPY	75	50	100	87.5
ROTIN	50	70	95	85
ELTTP	50	80	95	90
2nd run:	II-117 (75)	C (100)	II-117 + C (75 + 100)	II-117 + C
LIDPY	70	60	90	88
3rd run:	II-122 (60)	D (40)	II-122+D (60+40)	II-122+D
ECHSS	0	80	85	80
SCPSS	60	30	80	72
LIDPY	70	40	90	82
4th run:	II-18 (60)	E (300)	II-18+E (60+300)	II-18+E
MOOVP	80	60	100	92
LIDPY	60	60	95	84
CYPSE	50	40	80	70

Table 5:(Continued)

Test plant	Herbicidal efficacy (%)			Expected activity E according to Colby (%)
	compound(1) (g a.i./ha)	known herbicide (g a.i./ha)	compound(1) + known herbicide (g a.i./ha)	
5th run:	III-71 (125)	F (500)	III-71+F (125+500)	III-71+F
LIDPY	80	40	95	88
ROTIN	70	40	90	82
run6:	III-71 (125)	G (75)	III-71+G (125+75)	III-71+G
LIDPY	80	60	98	92
ROTIN	70	60	95	88

Test Example 6: Test for safening action on rice by water surface application

Test method

In a greenhouse, paddy soil was filled in pots (1,000 cm^2), and seeds of rice (cv. Nipponbare) were sown in the surface layer of the soil in the pots under wet conditions. 7 days after seeding, at the one leaf stage of the rice seedlings, the pots were watered to 3 cm in depth and the depth was kept during the test period. When the rice seedlings grew up to 1.5 leaf stage during the 9 days after seeding, a predetermined amount of the compound as a testing solution prepared in the same manner as the above-mentioned Test Example 4 was applied to the pots by water

surface treatment method. The phytotoxicity to rice seedlings was evaluated at 3 weeks after the application on a scale of 0 (no damage) to 100 (complete deth).

Test result of test example 6 are shown in Table 6.

Safening action of Test Example 6 were evaluated by Colby's equation.

$$\text{Colby}: E = X + \left[Y \times \frac{(100-X)}{100} \right]$$

E : expected phytotoxicity at p + q g/ha

X : expected phytotoxicity at p g/ha

Y : expected phytotoxicity at q g/ha

Compounds (1) in Table 6 are listed by by the compound numbers previously uesd in Tables 1, 2 and 3.

In Table 6 the known safeners are represented by the capital letters as shown in the following list:

a : N,N-diallyl-2,2-dichloroacetamide (dichlormid),

b : 4,6-dichloro-2-phenylpyrimidine (fenclorin),

c : diethyl (RS)-1-(2,4-dichlorophenyl)-5-methyl-2-pyrazoline-3,5-dicarboxylate (mefenpyr-diethyl),

d : N-(4-methylphenyl)-N'-(1-methyl-1-phenylethyl)urea (dymron),

e : 2-(dichloroacetyl)-2,2,5-trimethyl-oxazolidine (R-29148),

f : 1H,3H-naphtho [1,8-cd] pyran-1,3-dione (naphthalic anhydride).

Table 6: Safening efficacy (%) by water surface application

compound(1) (g a.i./ha)	Phyto-toxicity (%)	safener (g a.i./ha)	phytotoxicity (%)	compound (1) + safener (g a.i./ha)	phyto toxicity (%)	Expected phytotoxicity(E) according to Colby (%)
II-276 (400)	40	a (200)	0	II -276 + a (400 +200)	5	40
		b (400)	0	II -276 + b (400 +400)	20	40
		c (400)	20	II -276 + c (400 +400)	25	52
II-131 (400)	40	d (400)	0	II -131+ d (400 +400)	10	40
		b (400)	0	II -131+ b (400+200)	15	40
		e (200)	0	II -131 + e (400+200)	20	40
II-122 (600)	30	d (400)	0	II -122 + d (600 +400)	10	30
		a (200)	0	II -122 + a (600+200)	10	30
		b (400)	0	II -122 + b (600+400)	5	30
		f (400)	0	II -122 + f (600+400)	0	30
II-117 (400)	60	d (400)	0	II -117+ d (400 +400)	30	60
		a (200)	0	II -117+ a (400+200)	40	60
		f (400)	0	II -117+ f (400+400)	30	60
III-71 (600)	30	a (200)	0	III -71+ a (600 +200)	10	30
		c (400)	20	III -71+ c (600+400)	20	44

Table 6 : (Continued)

compound(1) (g a.i./ha)	Phyto-toxicity (%)	safener (g a.i./ha)	phytotoxicity (%)	compound (1) + safener (g a.i./ha)	phyto toxicity (%)	Expected phytotoxicity (E) according to Colby (%)
II-194 (400)	50	d (400)	0	II-194+ d (400 +400)	25	50
		b (400)	0	II-194+ b (400+400)	30	50
		f (400)	0	II-194+ f (400+400)	20	50

Formulation Example 1 (Granule)

To a mixture of the compound No. II-18 of the present invention (2.5 parts), bentonite (montmorillonite) (30 parts), talc (65.5 parts) and ligninsulphonate salt (2 parts), water (25 parts) is added. The mixture is well kneaded, made in granules of 10-40 mesh by an extrusion granulator and dried at 40-50°C to obtain a granule.

Formulation Example 2 (Granule)

Clay mineral particles having particle size distribution of 0.2-2 mm (95 parts) are put in a rotary mixer. While rotating it, the compound No. II-117 of the present invention (5 parts) is sprayed together with a liquid diluent into the mixer wetted uniformly and dried at 40-50°C to obtain granules.

Formulation Example 3 (Emulsifiable concentrate)

The compound No. II-122 of the present invention (30 parts), xylene (5 parts), poly-oxyethylenealkyl phenyl ether (8 parts) and calcium alkylbenzenesulfonate (7 parts) are mixed and stirred to obtain an emulsion.

Formulation Example 4 (Wettable powder)

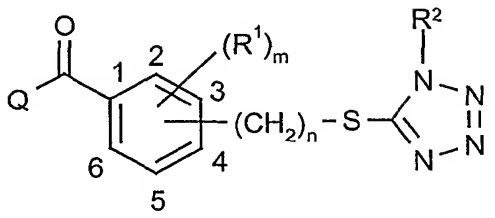
The compound No. II-194 of the present invention (15 parts), a mixture of white carbon (hydrous amorphous silicon oxide fine powders) and powder clay (1:5) (80 parts), sodium alkylbenzenesulfonate (2 parts) and sodium alkylnaphthalene-sulfonate-formalin-polymer (3 parts) are mixed in powder form and made into a wettable powder.

Formulation Example 5 (Water-dispersible granule)

The compound No. II-18 of the present invention (20 parts), sodium ligninsulfonate (30 parts), bentonite (15 parts) and calcined diatomaceous earth powder (35 parts) are well mixed, added with water, extruded using a 0.3 mm screen and dried to obtain a water-dispersible granules.

Claims

1. A compound of the formula (I)



wherein

R^1 represents halogen, methyl, ethyl, halomethyl, methoxy, ethoxy, C_{1-2} haloalkoxy, methylthio, ethylthio, C_{1-3} alkylsulfonyl, methylsulfonyloxy, ethylsulfonyloxy, nitro or cyano,

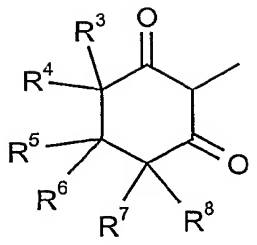
R^2 represents C_{1-6} alkyl, C_{3-6} cycloalkyl which may be optionally substituted with halogen or C_{1-3} alkyl, C_{1-4} haloalkyl, C_{2-6} alkenyl, or phenyl which may be optionally substituted with halogen, C_{1-3} alkyl, C_{1-2} haloalkyl or nitro,

m represents 0, 1 or 2,

the two R^1 substituents may be identical or different, in case m represents 2,

n represents 1 or 2,

Q represents one of the following groups



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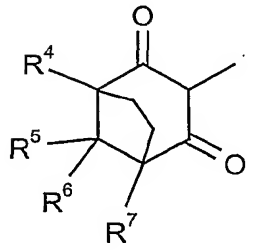

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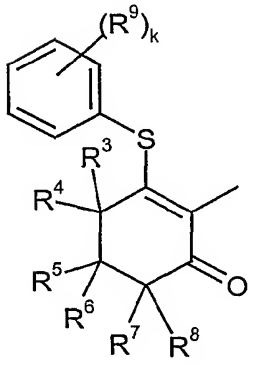

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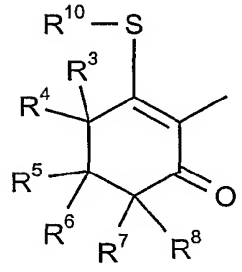

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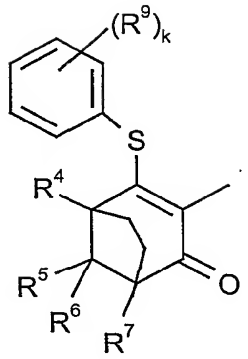

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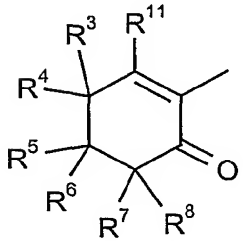
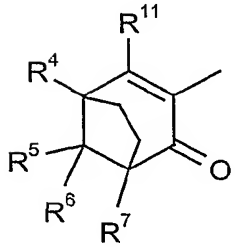


(Q-6),

or



(Q-7),

wherein

R^3, R^4, R^5, R^6, R^7 and R^8 are identical or different and each represents a hydrogen atom or methyl,

R^9 represents a hydrogen atom, halogen, C_{1-3} alkyl, halomethyl, methoxy or nitro,

R^{10} represents C_{1-6} alkyl,

R^{11} represents halogen, and

k represents 1 or 2.

2. A compound of the formula (I) according to Claim 1 wherein

R^1 represents fluoro, chloro, bromo, methyl, ethyl, trifluoromethyl, methoxy, ethoxy, C_{1-2} haloalkoxy, methylthio, ethylthio, methylsulfonyl, ethylsulfonyl, methylsulfonyloxy, ethylsulfonyloxy, nitro or cyano,

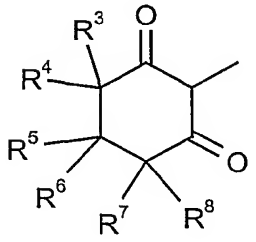
R^2 represents C_{1-3} alkyl, cyclopropyl which may be optionally substituted with fluoro, chloro, methyl or ethyl, C_{1-3} haloalkyl, C_{2-4} alkenyl, or phenyl which may be optionally substituted with fluoro, chloro, methyl, ethyl, trifluoromethyl or nitro,

m represents 1 or 2,

the two R^1 substituents may be identical or different, in case m represents 2,

n represents 1 or 2,

Q represents one of the following groups



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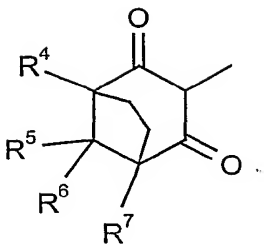

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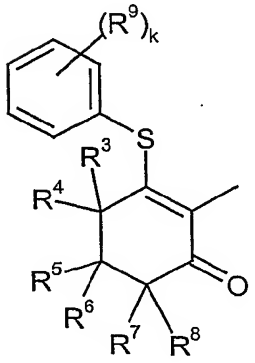

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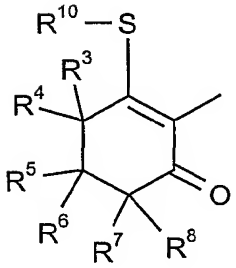

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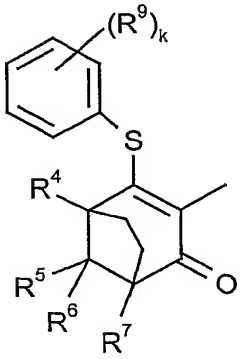
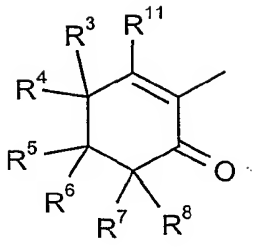
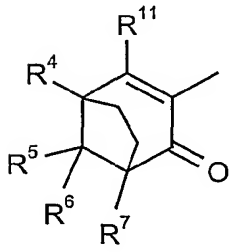


(Q-5),



(Q-6),

or



(Q-7),

wherein

R^3, R^4, R^5, R^6, R^7 and R^8 are identical or different and each represents a hydrogen atom or methyl,

R^9 represents a hydrogen atom, fluoro, chloro, methyl, ethyl or trifluoromethyl

R^{10} represents methyl or ethyl,

R^{11} represents chloro or bromo, and

k represents 1.

3. The compounds of the formula (I) according to in Claim 1 wherein

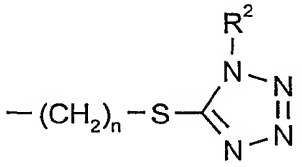
R^1 represents chloro, bromo, methyl or methylsulfonyl,

R^2 represents methyl, ethyl, n-propyl, isopropyl or cyclopropyl,

m represents 2, and in this case the two R^1 substituents are bond respectively to the 2-position and 4-position of a benzene ring and the two R^1 substituents may be identical or different,

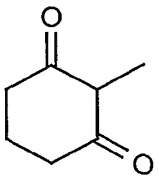
n represents 1,

the group



bonds to the 3-position of the benzene ring, and

Q represents one of the following groups



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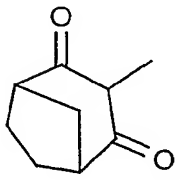
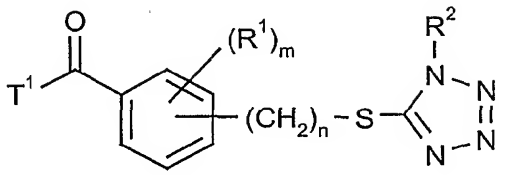


4. A process for the preparation of the compounds of Claim 1

characterized in that

a) in case of preparing a compound of the formula (I) wherein Q represents groups (Q-1) or (Q-2):

compounds of the formula (II)

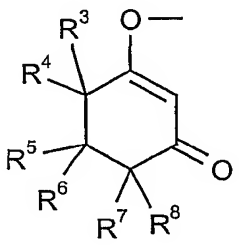


(II)

wherein

R^1, R^2, m and n have the same definition as mentioned in Claim 1, and

T^1 represents one of the following groups



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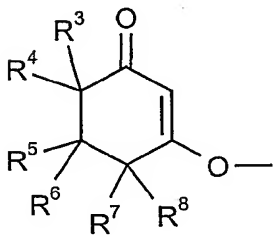

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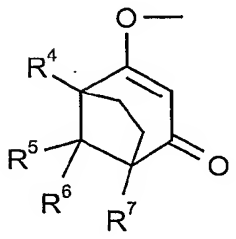

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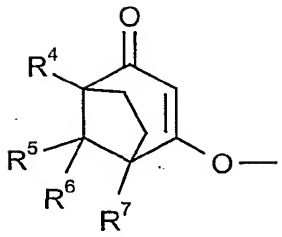
or



,

wherein

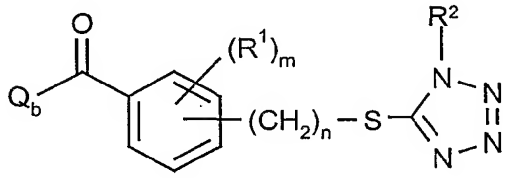
R^3, R^4, R^5, R^6, R^7 and R^8 have the same definition as mentioned in Claim 1,

are reacted to a rearrangement in the presence of inert solvents, and if appropriate, in the presence of a base and cyanide, and if appropriate, in the presence of a phase-transfer catalyst,

or

b) in case of preparing a compound of the formula (I) wherein Q represents groups (Q-6) or (Q-7) and R^{11} in said groups represents chloro or bromo:

compounds of the formula (Ib)

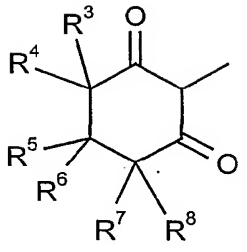


(Ib)

wherein

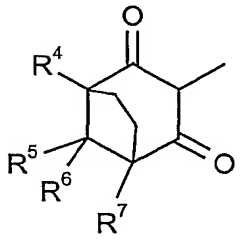
R^1, R^2, m and n have the same definition as mentioned in Claim 1, and

Q_b represents one of the following groups



(Q-1),

or



(Q-2),

wherein

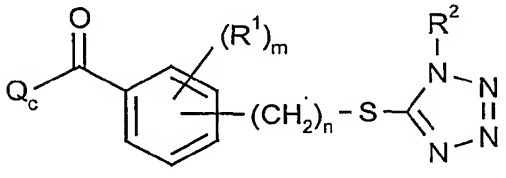
R^3, R^4, R^5, R^6, R^7 and R^8 have the same definition as mentioned in Claim 1,

are reacted with a halogenating agent in the presence of inert solvents,

or

c) in case of preparing a compound of the formula (I) wherein Q represents groups (Q-3), (Q-4) or (Q-5):

compounds of the formula (Ic)

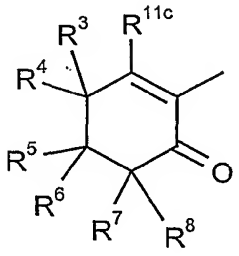


(Ic)

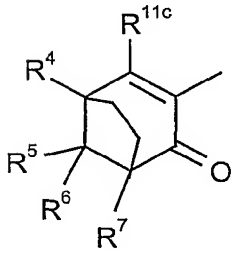
wherein

R^1, R^2, m and n have the same definition as mentioned in Claim 1, and

Q_c represents one of the following groups



or



wherein

R^3, R^4, R^5, R^6, R^7 and R^8 have the same definition as mentioned in Claim 1,

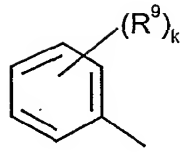
R^{11c} represents chloro or bromo,

are reacted with compounds of the formula (III)

$$R^{12}\text{-SH} \qquad \text{(III)}$$

wherein

R^{12} represents the group



or

R^{10}

wherein

R^9, R^{10} and k have the same definition as mentioned in Claim 1,

in the presence of inert solvents, and if appropriate, in the presence of an acid binding agent.

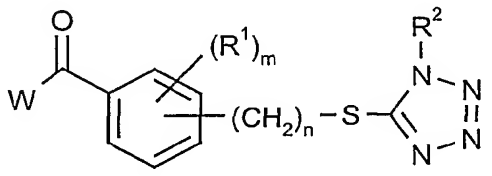
5. A herbicidal composition, characterized in that it contains at least one compound according to Claim 1.

6. A method for combating weeds, characterized in that a compound according to Claim 1 is allowed to act on weeds and / or their habitat.

7. Use of a compound according to Claim 1 for combating weeds.

8. A process for the preparation of herbicidal compositions, characterized in that a compound according to Claim 1 is mixed with extenders and / or surface active agents.

9. A compound represented by the formula (X)

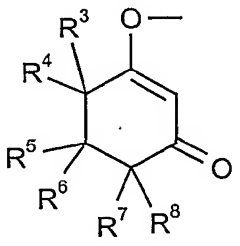


(X)

wherein

R^1, R^2, m and n have the same definition as mentioned in Claim 1,

W represents halogen, hydroxy, C_{1-4} alkoxy or one of the following groups



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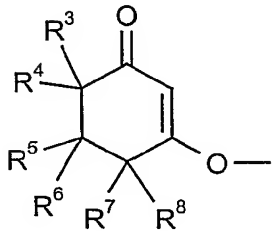

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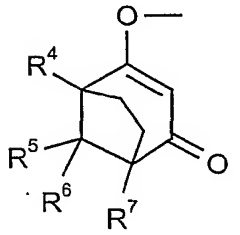

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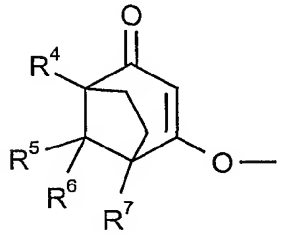
or



,

wherein

R^3, R^4, R^5, R^6, R^7 and R^8 have the same definition as mentioned in Claim 1.

10. A Herbicidal composition, characterized in that it contains an acitive substance combination comprising one tetrazole derivative of the formula (I) according to Claim 1 and at least one known active ingredient selected from the group consisting of acetamide type herbicides, amide type herbicides, benzofuran type herbicides, indanedione type herbicides, pyrazole type herbicides, oxazinone type herbicides, sulfonylurea type herbicides, thio-carbamate type herbicides, triazine type herbicides, triazole type herbicides, quinoline type herbicides, isoxazole type herbicides, dithiophosphate type herbicides, oxyacetamide type herbicides, tetrazolinone type herbicides, dicarboxyimide type herbicides, trione type herbicides, phenoxypropinate type herbicides, benzoic acid type herbicides, diphenylether type herbicides, pyridinedicarbothioate type herbicides, phenoxy type herbicides, urea type herbicides, naphthalenedione type herbicides and isoxazolidinone type herbicides.

11. A herbicidal composition, characterized in that it contains an active substance combination comprising one tetrazole derivative of formula (I) according to Claim 1 and at least one known safener selected from the group consisting of AD-67, BAS-145138, Benoxacor, Cloquintocet (-mexyl), Cyometrinil, 2,4-D, DKA-24, Dichlormid, Dymron, Fenclorim, Fenchlorazol (-ethyl), Flurazole, Fluxofenim, Furilazole, Isoxadifen (-ethyl), Mefenpyr (-diethyl), MG-191, Naphthalic anhydride, Oxabetrinil, PPG-1292, R-29148.

12. A Herbicidal composition, characterized in that they contain an active substance combination comprising one tetrazole derivative of the formula (I) according to Claim 1 together with at least one active ingredient selected from the group consisting of acetamide type herbicides, amide type herbicides, benzofuran type herbicides, indanedione type herbicides, pyrazole type herbicides, oxazinone type herbicides, sulfonylurea type herbicides, thio-

carbamate type herbicides, triazine type herbicides, triazole type herbicides, quinoline type herbicides, isoxazole type herbicides, dithiophosphate type herbicides, oxyacetamide type herbicides, tetrazolinone type herbicides, dicarboxyimide type herbicides, trione type herbicides, phenoxypropinate type herbicides, benzoic acid type herbicides, diphenylether type herbicides, pyridinedicarbothioate type herbicides, phenoxy type herbicides, urea type herbicides, naphthalenedione type herbicides and isoxazolidinone type herbicides, and at least one known safener selected from the group consisting of AD-67, BAS-145138, Benoxacor, Cloquintocet (-mexyl), Cyometrinil, 2,4-D, DKA-24, Dichlormid, Dymron, Fenclorim, Fenchlorazol (-ethyl), Flurazole, Fluxofenim, Furilazole, Isoxadifen (-ethyl), Mefenpyr (-diethyl), MG-191, Naphthalic anhydride, Oxabetrinil, PPG-1292, R-29148.

13. A process for combating weeds, characterized in that an active substance combination according to any of Claims 10, 11 or 12 is allowed to act on the weeds and/or their habitat.

14. Use of an active substance combination according to any of Claims 10, 11 or 12 for combating weeds.

15. A process for the preparation of a herbicidal composition, characterized in that an active substance combination according to any of Claims 10, 11 or 12 is mixed with extenders and/or surface-active agents.

INTERNATIONAL SEARCH REPORT

Inte ›nal Application No
PCT/IB 01/01130

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C07D257/04 A01N43/713

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C07D A01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)
EPO-Internal, PAJ, WPI Data, BEILSTEIN Data, CHEM ABS Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DE 198 46 792 A (HOECHST SCHERING AGREVO GMBH) 13 April 2000 (2000-04-13) cited in the application claims	1-15
A	WO 99 10327 A (DEYN WOLFGANG VON ;HILL REGINA LUISE (DE); RHEINHEIMER JOACHIM (DE) 4 March 1999 (1999-03-04) cited in the application claims	1-15
A	EP 0 103 143 A (BASF AG) 21 March 1984 (1984-03-21) claims	1-15

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

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"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
8 October 2001	16/10/2001

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31–70) 340–2040, Tx. 31 651 epo nl, Fax: (+31–70) 340–3016	Chouly, J

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INTERNATIONAL SEARCH REPORT

Information on patent family members

Inte[illegible]nal Application No
PCT/IB 01/01130

Patent document cited in search report		Publication date	Patent family member(s)		Publication date
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			BR	9914390 A	07-08-2001
			WO	0021924 A1	20-04-2000
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			EP	0103143 A2	21-03-1984
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